PB82-113028

The Evaluation of Alternatives for the Ultimate Disposal of Krypton-85
Task 1 - Compilation of Alernatives

Ford, Bacon and Davis Utah, Inc.
Salt Lake City

Prepared for

Environmental Protection Agency
Washington, DC

Jun 79

U.S. Department of Commerce
National Technical Information Service
NTIS

ORP Contract
Report 81-4

PB82-113028

THE EVALUATION OF ALTERNATIVES FOR THE ULTIMATE DISPOSAL OF KRYPTON-85

JUNE 1979

PREPARED FOR

U.S. ENVIRONMENTAL PROTECTION AGENCY
CONTRACT NO. 68-01-5001

BY

Ford, Bacon & Davis Utah Inc.

375 CHIPETA WAY SALT LAKE CITY UTAH 84108 · 801 · 583-3773

REPRODUCED BY
NATIONAL TECHNICAL
INFORMATION SERVICE
U.S. DEPARTMENT OF COMMERCE
SPRINGFIELD, VA 22161

NOTICE

This report was prepared by Ford, Bacon & Davis Utah, Inc., for the United States Environmental Protection Agency's Office of Radiation Programs (ORP) under Contract No. 68-01-5001. ORP has reviewed it, and the contractor has responded to our comments. We are publishing this report because of its useful information. We have not verified all of the results ourselves, however; nor have we applied our own editorial standards to the text. ORP does not necessarily publish all of the contractor reports it receives.

BIBLIOGRAPHIC DATA SHEET	1. Report No.	2.	3. Recipient's Accession No. PB82-113028

4. Title and Subtitle
The Evaluation of Alternatives for the Ultimate Disposal of Krypton-85

5. Report Date
June 1979

6.
U274018

7. Author(s)
B.J. Thamer, P.J. Macbeth, C.M. Mihlfeith, D.C. Rich

8. Performing Organization Rept. No.

9. Performing Organization Name and Address
Ford, Bacon & Davis Utah Inc.
375 Chipeta Way
P.O. Box 8009
Salt Lake City, Utah 84108

10. Project/Task/Work Unit No.

11. Contract/Grant No.
68-01-5001

12. Sponsoring Organization Name and Address
Office of Radiation Programs
U.S. Environmental Protection Agency
401 M St. SW
Washington, D.C. 20460

13. Type of Report & Period Covered

14.

15. Supplementary Notes

16. Abstracts

Task 1 reviews the literature on the immobilization and storage of krypton-85. In all, seventeen methods are reviewed, including steel cylinders at 500 and 2000 psi, zeolites, ion implantation/sputtering, krypton loaded solid set in a matrix, etc. A bibliography is provided for each technique. Task 2 compares twenty immobilization/storage techniques in terms of technical feasibility, safety, relative cost, and "other factors" which include retrievability, sociopolitical considerations, and transportational aspects. Two promising storage techniques (ion implantation and subatmospheric storage in tanks), along with the reference cases of 500 psi and 2000 psi storage in steel cylinders are considered with respect to ultimate disposal techniques in Task 3. The ultimate disposal techniques considered are storage in a building, storage in a dry well, storage in deep geological media, and ocean disposal. Specific designs are described in terms of achievability, environmental hazards, and comparative costs whenever possible.

17. Key Words and Document Analysis. 17a. Descriptors

Krypton-85, waste management, waste disposal

17b. Identifiers/Open-Ended Terms

17c. COSATI Field/Group

18. Availability Statement	19. Security Class (This Report) UNCLASSIFIED	21. No. of Pages
	20. Security Class (This Page) UNCLASSIFIED	22. Price

FORM NTIS-35 (REV. 10-73) ENDORSED BY ANSI AND UNESCO. THIS FORM MAY BE REPRODUCED USCOMM-DC 8265-P74

1<

THE EVALUATION OF ALTERNATIVES
FOR THE ULTIMATE DISPOSAL OF KRYPTON-85

TASK 1 - COMPILATION OF ALTERNATIVES

FEBRUARY 1979

prepared for

U.S. ENVIRONMENTAL PROTECTION AGENCY

CONTRACT NO. 68-01-5001

by

B. J. Thamer
C. M. Mihlfeith
P. J. Macbeth

FORD, BACON & DAVIS UTAH INC.

375 CHIPETA WAY

P.O. BOX 8009

SALT LAKE CITY, UTAH 84108

CONTENTS

Chapter Title Page

List of Figures v
List of Tables. vi
Abstract. vii

1 INTRODUCTION

1.1 Background 1-1
1.2 Scope and Objectives 1-2
1.3 Methodology. 1-3
References for Introduction 1-6
Bibliography for Introduction. 1-10

2 THE COMPILATION OF POSSIBLE TECHNIQUES

2.1 Gas at 500 or 2000 psi in a Cylinder 2-1
References 2-4
2.2 Zeolites 2-7
References 2-10
2.3 Krypton-Loaded Solid Set in a Matrix 2-12
References 2-16
2.4 Ion Implantation/Sputtering. 2-17
References 2-20
Bibliography 2-23
2.5 Metal Vapor Deposition 2-25
References 2-26
Bibliography 2-27
2.6 Clathrates 2-28
References 2-31

3<

CONTENTS (cont)

Chapter Title Page

Bibliography 2-32

2.7 Porous Underground Media 2-33

References 2-37

2.8 Hydrofracture. 2-39

References 2-40

2.9 Extraterrestrial 2-43

References 2-44

Bibliography 2-45

2.10 Disposal in the Ocean. 2-47

References 2-49

2.11 Storage on Adsorbents. 2-50

References 2-52

2.12 Transmutation. 2-55

References 2-59

Bibliography 2-60

2.13 Cryogenic Storage. 2-61

References 2-62

2.14 Low-Pressure Storage in Caverns. 2-63

References 2-65

2.15 Low-Pressure Storage in Tanks. 2-66

References 2-68

2.16 Others 2-69

2.16.1 Solutions. 2-69

References for Solutions 2-71

Bibliography for Solutions 2-73

4<

CONTENTS (cont)

Chapter	Title	Page
	2.17 Combinations of Alternatives	2-74
	References	2-75
3	SUMMARY OF ALTERNATIVE TECHNIQUES	3-1

APPENDIX A - TABLES AND GLOSSARY

LIST OF FIGURES

Number	Title	Page
2.2.1	Process for Loading Krypton into Sorbent. . . .	2-8
2.4.1	Experimental Diagram for Ion Implantation/ Sputtering.	2-18
2.12.1	The Relative Time in a PWR Flux	2-57

6<

LIST OF TABLES

Number	Title	Page
2.3.1	Comparison of Candidate Processes for Solid Storage	2-13
2.3.2	Some Metals and Alloys for a Possible Matrix	2-14
2.3.3	Advantages and Disadvantages of Krypton-Loaded Solid Set in a Matrix	2-15
2.6.1	Advantages and Disadvantages of Quinol Clathrates	2-30
2.7.1	Criteria for Porous Media of an Underground Storage Site	2-34
2.7.2	Advantages and Disadvantages of Porous Underground Media	2-36
2.10.1	Advantages and Disadvantages of Disposal in the Ocean	2-48
2.11.1	Advantages and Disadvantages of Krypton Adsorbed on Charcoal	2-51
2.14.1	Advantages and Disadvantages of Low-Pressure Storage in Caverns	2-64
2.15.1	Advantages and Disadvantages of Storage in Low-Pressure Tanks	2-67
2.16.1	Advantages and Disadvantages of Organic Solutions	2-70

7<

ABSTRACT

A survey of the literature is presented on the possible means of immobilizing krypton-85, an inert fission product gas that poses a radiological hazard if released to the atmosphere. The literature on disposal and storage of krypton has been reviewed and summarized in Chapter 2. The alternative methods are categorized into the following principal techniques: (1) 500 or 2000 psi gas, (2) zeolites, (3) loaded solid set in a matrix, (4) ion implantation/sputtering, (5) metal vapor deposition (6) clathrates, (7) porous underground media, (8) hydrofracture, (9) extraterrestrial disposal, (10) disposal in the ocean, (11) adsorbents (12) transmutation, (13) cryogenics, (14) low-pressure storage in caverns, (15) low-pressure storage in tanks, (16) solutions, and (17) combinations of the foregoing. A bibliography is given for each technique. Chapter 3 contains a summary of the available body of knowledge on each technique. The techniques are to be evaluated in more detail in subsequent tasks of this project.

8<

1. INTRODUCTION

1.1 BACKGROUND

The International Nuclear Fuel Cycle Evaluation (INFCE) program is in progress, and alternatives to commercial reprocessing of nuclear fuel may be recommended; nevertheless, commercial reprocessing is still actively being considered on the international scene and conceivably might have a future use.[1] Furthermore, noncommercial reprocessing may continue on some scale. The atmospheric testing of nuclear weapons has contributed to the atmospheric concentrations of krypton-85, but these tests no longer occur on a large scale. Commercial reprocessing is thus potentially the principal continuing source of environmental pollution by krypton-85, although minor amounts (<<1% of the amount produced) also are released during the normal operation of light-water reactors (LWR's).[2] Small amounts also could be involved in beneficial uses of krypton-85.

Krypton is a rare gas that can form chemical compounds, but only with difficulty. A fuel reprocessing plant produces elemental gaseous krypton usually having an isotopic composition of about 6 % krypton-85.[3] This radioactive krypton mixture generally has been released to the atmosphere through a stack. Exposures to individuals several kilometers downwind of the release can be estimated by well known methods.[4,5,6] The radioactivity persists in the atmosphere due to the 10.73-year half life of the krypton-85 (or ^{85}Kr). This radioactivity becomes uniformly distributed throughout the surface air over the earth within about 2 years.[7] Although the solubility of krypton in pure water[8] and sea water[9] is appreciable, the loss of krypton from the air to surface waters is probably less than 2%,[10] and the loss to washout and other deposition is probably nil.[11] Consequently the undecayed krypton radioactivity remains essentially in the air.

Environmental analyses have been made concerning the possible releases of krypton-85 from the fuel cycle.[12,13] One such analysis has been made for a generic fuel reprocessing plant envisioned to start in 1983 and run for 20 years.[13] The projected growth rate of the world population is consistent with the recent value of 4.3 billion persons.[14] The 1976 analysis for a plant of 2100 metric tons of fuel per year and an offgas flow rate of 50 SCFM[13] has been modified (pro rata) here to 2240 metric tons per year. It is assumed that 99% of the krypton-85 is prevented from entering the atmosphere during the 90% of the time that the krypton reprocessing plant is in operation, but that the krypton-85 is put up the stack during the remaining 10% of the time. One estimates the world-wide commitment averted by the storage of the krypton-85 from the 0.99 x 0.90 x 2240 = 2000 metric tons per year for 20 years to be as follows (in man-kilorem): whole body, 199; gonads, 266; and lungs, 399. This results in the averting of 149 health

9<

effects (in terms of cancer induction and/or mortality). The reprocessing costs for the krypton-85 are estimated[13] to be rather small compared to NRC's interim value of $1,000 per whole-body man-rem.[15]

The global krypton-85 inventory in the atmosphere is already quite sizeable as a result of past tests of nuclear weapons and of fuel processing for the production of plutonium.[10] Sufficient atmospheric krypton-85 may exist or be produced to alter significantly the electrical fields of the atmosphere and thus affect the weather.[16] On January 13, 1977 the radiation-protection standard promulgated by the U.S. Environmental Protection Agency was published in the Federal Register to the effect that the krypton-85 entering the atmosphere from the fuel cycle should not exceed 50,000 Ci per gigawatt-year of electrical energy.[17] The rule is to be effective January 1, 1983, even though commercial reprocessing does not exist at the present time. The above figure requires the reduction of the evolution of krypton-85 to about 14 % of its previous value. Thus the quantitative removal and disposal of krypton-85 is not to be required even by 1983. At the present time, disposal techniques might be considered on the basis of possibly less than complete removal, although requirements at a future date might become more stringent.

The krypton from the reprocessing of 2000 metric tons of fuel has been quantified by Knecht[3] and Foster et al.[18] as shown in the following summary for 28,700 MWd burnup/ton:

6 volume % ^{85}Kr

17,000,000 Ci ^{85}Kr,

191 m^3 (STP) of total Kr,

99% of decay energy from 0.67 MeV beta decay,

24.7 kW of decay heat at the beginning of its decay with a half life of 10.73 years

The above data might be taken as typical of the annual production from 67 reactors of approximately 1000 MWe capacity each. If appreciable xenon, for example, also were present, the volume of gas to be handled would be changed accordingly. (Appendix A contains physiochemical data on krypton and xenon.)

1.2 SCOPE AND OBJECTIVES

The scope of Task 1 of this study is the presentation of possible alternatives for the disposal of krypton-85. The ultimate purpose of the compilation is to aid the EPA in preserving the quality of the environment at a reasonable cost.

10<

The storage of radioactive krypton gas in cylinders at 500-2000 psi (3.4-13.6 MPa) serves as the reference against which other alternatives may be compared because of the greater experience with the gas-storage technique. This study examines the reference case and all feasible alternative techniques. Basic technologies are presented rather than the details of each or the systems relationships. More detailed aspects are to be examined in later tasks after the more viable alternatives have been selected for evaluation.

The radioactive krypton-85 is assumed to be supplied in the elemental, gaseous form. The gas may be assumed to be obtained at a fuel reprocessing plant where a process for separating krypton has been installed.[19] The latter may be a method of cryogenic distillation (producing krypton nearly free of xenon)[20-22] or a method of solvent extraction using CCl_2F_2, for example.[23-26] The latter method produces the krypton admixed with a larger volume of the rare gas xenon, but it may have fewer other impurities.

The effect of the xenon and other gases on the concentration of stored kryption-85 is clear enough for some alternatives, such as the storage of gas in cylinders at a fixed total pressure, but its effect is less obvious in some other cases. An attempt is made in Chapter 2 to comment on this aspect for the latter cases to the extent that information is available.

1.3 METHODOLOGY

This compilation has been performed by consulting the written literature, the data bases of computer programs, and outstanding personnel in the field. A previous categorization of alternatives for the disposal of low-level radioactive waste (LLW) has been adaptable in uncovering and classifying methods for the disposal of krypton-85.[27] Thus, significant technologies in such diverse areas as extraterrestrial disposal, atmospheric disposal, disposal to waters, disposal in the earth, disposal in engineered structures, and disposal by transmutation have been found.

Extensive use has been made of the scientific libraries of the Idaho National Engineering Laboratory and the University of Utah. The following documents have been especially helpful as sources of information:

(a) "Alternatives for Managing Wastes from Reactors and Post-Fission Operations in the LWR Fuel Cycle," Report ERDA-76-43, Vol 2, Section 13.1,[19] and Section 14.1,[28] May 1976

(b) Proceedings from the last several Air Cleaning Conferences[29]

11<

(c) Proceedings of the International Symposium on the Management of Wastes from the LWR Fuel Cycle held in Denver, Colorado, July 11-16, 1976[30]

(d) Certain documents from the Idaho National Engineering Laboratory, e.g. one in 1977[3] and another in 1973[31]

(e) A 1976 paper from Argonne National Laboratory[32]

Also undertaken in the survey were manual and computer searches at the two libraries. The searches covered the literature on krypton-85, including the following sources:

(a) DOE Energy Database (TIC), 1974-Oct 1978 (incl)

(b) Nuclear Science Abstracts, 1967-June 1976 (incl)

(c) Atomindex, 1976-Sept 1978 (incl)

(d) National Technical Information Service, 1970-Oct 1978 (incl)

(e) Chemical Abstracts, 1970-Sept 1978 (incl)

(f) Energy Research Abstracts, 1977-Sept 1978 (incl)

(g) Government Reports Announcements, 1968-1977 (incl)

(h) Scientific and Technical Aerospace Reports, 1963-June 1977 (incl)

(i) Smithsonian Science Information Exchange, Current Research, 1975-Aug 1978 (incl)

(j) Compendex, 1970-Oct 1978 (incl)

(k) Environline, 1971-Aug 1978 (incl)

(l) Pollution Abstracts, 1970-77 (incl)

(m) Environmental Periodicals Bibliography, 1974-Aug 1978

(n) APTIC (Air Pollution), 1966-Aug 1978 (incl)

(o) International Atomic Energy Agency to 1977 (incl)

In addition, the authors are acquainted with the personnel and facilities of the U.S. centers of research on the disposal of krypton-85.

All but the most recent reports of the national laboratories should be available at small cost from the National Techni-

12<

cal Information Service, U.S. Department of Commerce, 5285 Port Royal Road, Springfield, Virginia 22161.

13<

REFERENCES FOR INTRODUCTION

1. S. Rippon, "La Hague: French Face Bright Prospects for Commercial Oxide Fuel Reprocessing," Nuclear News 21 (14), 58 (1978); "Windscale - Now To Get On and Build It," Nuclear News 21 (15), 45 (1978).

2. "Krypton-85 in the Atmosphere-Accumulation, Biological Significance, and Control Technology," NCRP Report 44, National Council on Radiation Protection and Measurements, 7910 Woodmount Ave., Washington, D.C. 20014, July 1, 1975.

3. D.A. Knecht, "An Evaluation of Methods for Immobilizing Krypton-85," Report ICP-1125, National Technical Information Service, U.S. Department of Commerce, Springfield, Va. 22161, 1977.

4. D.H. Slade, ed., "Meteorology and Atomic Energy, 1968," Report TID-24190, National Technical Information Service, U.S. Department of Commerce, Springfield, Va. 22161, 1968.

5. P.W. Nickola, "A Comparison of Exposures Resulting from Inert Gas Releases with Exposures Predicted with the Aid of Pasquill's Diffusion Categories," p 123 in C.L. Simpson, Pacific Northwest Laboratory Annual Report for 1971, Vol II: Physical Sciences; Part 1, Atmospheric Sciences, Report BNWL-1651 PTI, Battelle Pacific Northwest Laboratories, Richland, Wash. 99352, Dec 1972.

6. D.L. Strange et al., "Review of Calculational Models and Computer Codes for Environmental Dose Assessment of Radioactive Releases," Report BNWL-B-454, June 1976.

7. L. Machta et al., "Regional and Global Scale Dispersion of ^{85}Kr for Population Dose Calculations," p 411 in Physical Behavior of Radioactive Contaminants in the Atmosphere, Symposium held in Vienna, Nov 12-16, 1973, International Atomic Energy Agency, Vienna, Austria, 1974.

8. J. Mishima and L.C. Schwendiman, "Some Observations on Noble Gas Partitioning in Dynamic Air-Water Systems," for presentation at the 11th AEC Air Cleaning Conference, Richland, Wash., Aug 31-Sept 3, 1970, Battelle Memorial Institute, Report CONF-700816-5, Battelle Pacific Northwest Laboratories, Richland, Wash., Aug 25, 1970.

9. D. Wood and R. Caputi, "Solubilities of Krypton and Xenon in Fresh and Sea Water," Report USNRDL-TR-988, U.S. Naval Radiological Defense Laboratory, San Francisco, Calif., Apr 1966.

14<

10. W.S. Diethorn and W.L. Stockho, "The Dose to Man from Atomospheric ^{85}Kr," Health Physics 23, 653 (1972).

11. J. Tadmor, "Deposition of ^{85}Kr and Tritium Released from a Nuclear Fuel Reprocessing Plant," Health Physics 24, 37 (Jan 1973).

12. U.S. Environmental Protection Agency, "Environmental Analysis of the Uranium Fuel Cycle, Part III, Nuclear Fuel Reprocessing," Report EPA-520/9-73-003-D, Office of Radiation Programs, Environmental Protection Agency, Washington, D.C. 20460, Oct 1973.

13. U.S. Environmental Protection Agency, "Environmental Analysis of the Uranium Fuel Cycle, Part IV, Supplementary Analysis-1976," Report EPA 520/4-76-017, Office of Radiation Programs, Environmental Protection Agency, Washington, D.C. 20460, July 1976.

14. The Wall Street Journal, Nov 20, 1978, p 40, The Wall Street Journal, 17 Inverness Way East, Englewood, Colo. 80112.

15. U.S. Nuclear Regulatory Commission, "Opinion of the Commission": In the Matter of Rulemaking Hearing, Numerical Guides for Design Objectives and Limiting Conditions for Operation to meet the Criterion, "As Low As Practicable" for Radioactive Material in Light-Water-Cooled Nuclear Power Reactor Effluents, Docket No. RM-50-2 (May 5, 1975).

16. W.L. Boeck, "Environmental Consequences of Atmospheric Krypton-85," Report COO-4364-1, National Technical Information Service, U.S. Department of Commerce, Springfield, Va. 22161, 1978.

17. U.S. Environmental Protection Agency, "Environmental Radiation Protection Standards for Nuclear Power Operations," Federal Register 42, No. 9, Title 40, Part 190 (1977).

18. B.A. Foster et al., "Long Term Storage Techniques for ^{85}Kr," p 293 in M.W. First, ed., Report CONF-740807-P1, Proceedings of the Thirteenth AEC Air Cleaning Conference, San Francisco, Calif., Aug 12-15, 1974.

19. U.S. Energy Research and Development Administration, "Alternatives for Managing Wastes from Reactors and Post-Fission Operations in the LWR Fuel Cycle," Report ERDA-76-43, Vol 2, Section 13.1, National Technical Information Service, U.S. Department of Commerce, Springfield, Va. 22161.

20. J.S. Davis and J.R. Martin, "A Cryogenic Approach to Fuel Reprocessing Gaseous Radwaste Treatment," p 176, Transac-

tions of the American Nuclear Society 1973 Meeting, Chicago, Ill., June 10-14, 1973.

21. H. Yusa et al., "Application of Cryogenic Distillation to Krypton-85 Recovery," Nuclear Engineering and Design 41, 437 (1977).

22. R.v. Ammon et al., "Development of a Cryogenic Krypton-Separation System for the Off Gas of Reprocessing Plants," presented at the 15th DOE Nuclear Air Cleaning Conference, Boston, Mass., Aug. 7-10, 1978.

23. M. Steinberg and B. Manowitz, "Recovery of Fission Product Noble Gases," Industrial Engineering Chemistry 51 (1), 47 (1959).

24. J.R. Merriman et al., "Removal of Radioactive Krypton and Xenon from Contaminated Off-Gas Streams," p 175 in M.W. First and J.M. Morgan, Jr., eds., Proceedings of the Eleventh AEC Air Cleaning Conference, Richland, Wash., Aug 31-Sept 3, 1970, Report CONF 700816, Vol 1, Dec 1970.

25. R.F. Taylor and G.P. Wall, "Development of a Production Process for Radio-Krypton Recovery by Fractional Absorption," p 307 in C.M. Nicholls, ed., Technology, Engineering and Safety, Vol 5 of Progress in Nuclear Energy, Series IV, 1963.

26. T.M. Gilliam et al., "Krypton Absorption in Liquid CO_2 (KALC): Campaign III in the Experimental Engineering Section Off-Gas Decontamination Facility," Report ORNL/TM-5655, National Technical Information Service, U.S. Department of Commerce, Springfield, Va., Sept 1977.

27. P.J. Macbeth et al., "Screening of Alternative Methods for the Disposal of Low-Level Radioactive Wastes," Report NUREG/CR-0308, Fig. 2.3, National Technical Information Service, U.S. Department of Commerce, Springfield, Va. 22161.

28. U.S. Energy Research and Development Administration, "Alternatives for Managing Wastes from Reactors and Post-Fission Operations in the LWR Fuel Cycle," Report ERDA-76-43, Vol 2, Section 14.1 National Technical Information Service, U.S. Department of Commerce, Springfield, Va. 22161.

29. Dr. M.W. First, ed.: 15th Air Cleaning Conference, Boston, Mass., Aug 7-10, 1978, Harvard Air Cleaning Laboratory, 665 Huntington Ave., Boston, Mass. 02115; Proceedings of the 14th ERDA Air Cleaning Conference Held in Sun Valley, Idaho, Aug 2-4, 1976, Vols 1 and 2, Reports CONF-760822-P1 and P2, NTIS, Feb 1977; Proceedings of the 13th AEC Air Cleaning Conference Held in San Francisco, Calif., Aug

16<

12-15, 1974, Vols 1 and 2, Reports CONF-740807-P1 and P2, NTIS, Mar 1975; Proceedings of the 11th AEC Air Cleaning Conference Held in Richland, Wash., Aug 31-Sept 3, 1970, Vol 1, Report CONF-700816 (Vol 1) NTIS, Dec 1970.

30. Energy Research and Development Administration, Proceedings of the International Symposium on the Management of Wastes from the LWR Fuel Cycle Held in Denver, Colo., July 11-16, 1976, Report CONF-76-0701, National Technical Information Service, U.S. Department of Commerce, Springfield, VA 22161.

31. C.L. Bendixsen and J.A. Buckham, "General Survey of Techniques for Separation and Containment of Noble Gases from Nuclear Facilities," Paper CONF-730915-16 in Symposium on Noble Gases, Las Vegas, Nev., Sept 24-8, 1973.

32. M. Steindler and B. Kullen, "Preparation of Collected Radionuclide Wastes for Storage," p 6.1-6.24 in Controlling Airborne Effluents from Fuel Cycle Plants, Proceedings of Topical ANS Meeting, Sun Valley, Idaho, Aug 5-6, 1976, American Nuclear Society, Hinsdale, Ill.

BIBLIOGRAPHY FOR INTRODUCTION

Anon, "Krypton, A DDC Bibliography, July 1961, to February 1969," Defense Documentation Centers, Alexandria, Va., 1969.

J.R. Berreth, "Fabrication of Radioactive Samples for Cross Section Measurements", Nuclear Applications 1, 230-234 (1965).

C.O. Bopp and O. Sismon, "Radiation Stability of Plastics and Elastomers", Report ORNL-1373, Oak Ridge National Laboratory, Oak Ridge, Tenn., 1953.

W.W. Brandt, "Solubilities and Diffusion Coefficients of Inert Gases", Phys. Stat. Sol. A-30, 263-269 (1975).

C.A. Burchsted et al., "Nuclear Air Cleaning Handbook. Design, Construction and Testing of High-Efficiency Air Cleaning Systems for Nuclear Application," Report ERDA-76-21, Oak Ridge National Laboratory, Oak Ridge, Tenn., 1976.

R.H. Burns, "Solidification of Low and Intermediate Fuel Wastes", Atomic Energy Review 9, 547-599 (1971).

W.E. Clark and R.E. Blanco, "Encapsulation of Noble Fission Product Gases in Solid Media Prior to Transportation and Storage", Report ORNL-4473, Oak Ridge National Laboratory, Oak Ridge, Tenn., Feb 1975.

J.J. Cohen and K.R. Peterson, "Considerations in Siting Long-Term Radioactive Noble Gas Storage Facilities", Paper presented at Noble Gas Symposium, Las Vegas, Nev., 1973.

J.R. Coleman and R. Liberace, "Nuclear Power Production and Estimated ^{85}Kr Levels," Radiological Health Data and Reports 7, 615-621 (1966).

D. Cuaz, A. Limongi, and A. Traxler, "Embedding Wastes from a Light Water Reactor in Thermosetting Resins", Transactions of the American Nuclear Society 20, 654-656 (1975).

J.A. Franz and L.L. Burger, "Polymer Media for Tritium Fixation", Report BNWL-B-430, Battelle-Pacific Northwest Laboratories, Richland, Wash., May 1975.

J.H. Holloway, "Noble-Gas Chemistry," Methuen and Co., Ltd., London, 1968.

W.W. Kellog and M. Mead, eds., "The Atmosphere: Endangered and Endangering," Fogarty International Center Proceedings No. 39, Oct 26-29, 1975, U.S. Gov. Printing Off. Wash., D.C. 20402 (Stock No. 017-053-00059-1).

18<

U.V. Kulichenko and I.A. Sobolev, "Properties of Cement Blocks and Definition of Conditions for Safe Disposal", USSR State Committee on the Utilization of Atomic Energy, Report 69-5207 (1969).

M. Laser, "Separation, Storage and Disposal of Krypton-85, Status and Projects," Report GERHTR-177, paper for the IAEA Technical Committee on Removal, Storage and Disposal of Gaseous Radionuclides from Airborne Effluents, Sept 1976.

K. Nakasima, "Management of Fuel Reprocessing Wastes in Japan", CONF-721107, pp 299-312.

E. Paul and G. Gehrke, "Krypton Physical and Physiochemical Characteristics, A Bibliography," Report JUL-BIBL.-11, Kernforschungsanlage, Juelich, West Germany, Sept 1966.

R.D. Penzhorn, "Alternate Processes for the Long-Term Storage of Kr-85," Report KFK-2482, Kernforschungszentrum Karlsruhe G.m.b.H., Germany, F.R. (in German), Dec 1977; Atomindex 9, 396106 (1978).

I. Ya Petrov and V.L. Korpov, "Investigation of Gas Evaluation from the Action of Nuclear Radiation on Polymers", Proceedings of 1st All-Union Conference on Radiation Chemistry, Moscow, 1957, Consultants Bureau, New York, N.Y., pp 263-268, 1959.

T.R. Pinchback and D.A. Knecht, "The Development of Process and Storage Materials Suitable for Krypton-85 Waste Management", paper presented at International Symposium on Science Underlying Radioactive Waste Management, Boston, Mass., Nov 29 - Dec 1, 1978.

Y. Sousselier, "Conditioning of Wastes from Power Reactors", Transactions of the American Nuclear Society 17, 332 (1973).

M. Steinberg, P. Colombo and B. Manowitz, "Concrete-Polymer Materials Applied to Long-Term Storage of Radioactive Waste", Transactions of the American Nuclear Society 17, 333-334 (1973).

D.L. Strange et al., "Review of Calculational Models and Computer Codes for Environmental Dose Assessment of Radioactive Releases," Report BNWL-B-454, Battelle Pacific Northwest Laboratories, Richland, Wash., June 1976.

G.V. Ulrickson, "Information Support of Energy Research and Development Administration's Environmental Program at Oak Ridge National Laboratory," Report CONF-750967-10, of 3rd ERDA Environmental Protection Conference, Chicago, Ill., Sept 23-26, 1975.

U.S. Energy Research and Development Administration, "Alternatives for Managing Wastes from Reactors and Post-Fission Operations in the LWR Fuel Cycle," Report ERDA-76-43, Vol 2, pp

13.9-13.11 and 14.6-14.7, National Technical Information Service, U.S. Department of Commerce, Springfield, Va. 22161, 1976.

S. Walters, "Nuclear Waste Storage", Mechanical Engineering 99, 63 (1977).

E.J. Weber and H. Heusinger, "Schutzeffek in Styrol-Acrynitril Copolymer bei der Einweikieng Ionisierender Strahlung", Radiochem Acta 4, 92-100 (1965).

20<

2. THE COMPILATION OF POSSIBLE TECHNIQUES

The available body of knowledge concerning the storage of krypton-85 has been examined. The intent has been to review all basic methods that have rational promise. Chapter 3 contains a brief summary of each technique described in the following sections.

2.1 GAS AT 500 OR 2000 PSI IN CYLINDERS

For over 20 years the Idaho National Engineering Laboratory and its predecessor have extracted krypton-85 from the reprocessing of nuclear fuel[1,2] and bottled it in nominally 50-liter, high-pressure steel cylinders for shipment to Oak Ridge National Laboratory in a special shipping cask.[3] The cylinder has an outside diameter of 9 in. and a height of 52 in.; it is a modified standard compressed-gas cylinder (DOT Specification 3A-2015). The valve has been modified to have a valve seat of monel and a valve stem of Type 303 stainless steel. The cylinder has been shipped with 500 psi or less of the product krypton containing also oxygen, nitrogen, xenon, water vapor and possibly NO_x from fuel reprocessing. The purity of this gas and its possible corrosive behavior would change with improvements in the method of preparation by cryogenic distillation[4-6] or by using another process such as solvent extraction.[7-8] The cylinder is tested at 3360 psig at least every 5 years and is certified for a pressure of 2015 psig.[3]

Low-alloy steel (typically A.I.S.I 4130) cylinders such as the one described above have been used for the storage and handling of industrial gases for over 50 years.[9] The gases have included such highly toxic ones as phosgene and arsine, but they have not been intensely radioactive. A cylinder could last at least 500 years if it suffered only normal exterior rusting and no abuse according to Union Carbide Corporation's study of sequential 5-year tests on several thousand cylinders.[9,10]. Restrictions on the shipment of krypton-85 at the Idaho National Engineering Laboratory have required that (a) the initial cylinder pressure not exceed 500 psi, (b) the heat generation be 293 watts (74,000 Ci) or less,[3,11] and (c) the cylinder be suitably shielded against gamma exposure.[12] The 500-psi limit presumably would not apply if the bottled krypton were stored at the site of the reprocessing plant. Foster et al have estimated the wall temperature of the 50-liter bottle with air cooling and a krypton pressure of approximately 500 or 2000 psi.[9,10] With an assumed 6% ^{85}Kr in the gas and an ambient air temperature of 21° C, 500 psi gives a 60° C wall temperature and 128,000 Ci per cylinder compared with a 127° C wall temperature and 419,000 Ci per cylinder for 2000 psi. These figures imply about 29 cm^3 (STP) of contained krypton per cm^3 of cylinder gas for 500 psi and 100 cm^3 (STP)/cm^3 for 2000 psi, respectively. The gas storage of krypton-85 at 500 psi has been prescribed for a reference Krypton Storage Facility.[13]

Even though the containment of krypton-85 in gas cylinders is the best developed alternative thus far, questions still remain on its reliability for a period of several decades. For example, the rubidium daughter of the decaying krypton-85 is a possible corrodent toward the cylinder and valve materials as are the gaseous oxygen, moisture and NO_x impurities in a high-radiation field. The impurities would convert the rubidium metal into an equivalent amount of rubidium oxide or hydroxide with accompanying corrosive effects. The Idaho National Engineering Laboratory is conducting tests on general corrosion, pitting, intergranular attack, embrittlement and the effects of gas purity as well as radiation.[14-16] The candidate materials include stainless steels and nickel base alloys in addition to 4130 steel. Valve-seat candidates probably do not include plastics because of their susceptibility to radiation damage as in the case of polytetrafluoroethylene and micarta.[9] Tests of up to 400° C for 5000 hr in liquid rubidium on stressed samples showed no liquid embrittlement or corrosion except for pitting on 4130 steel and on an unstabilized stainless steel (304). Monel 400, alloy A286, and two other stainless steels (316 and 347), for example, remained untouched. Hence pure rubidium metal has given no sign of intractable materials problems. However, a more oxided sample of rubidium at higher temperatures has given significant intergranular attack together with somewhat less serious general attack.[14] These and previous observations without rubidium suggest the need for further compatibility studies with gaseous impurities. The ultimate answer will bear on the probability of failure of a cylinder or its valve. Some estimates of Foster and Pence indicate that even if a 2000-psi cylinder of krypton-85 were suddenly to fail, the rate of dosage to someone in the facility would not necessarily be fatal.[9] Furthermore, such exposures likely would be precluded by remote operation and secondary containment.

A critical area may be the valve where the combined effects of mechanical wear, impurities and corrosion come to a focus at the valve seat. The valve and its brass plug have been closed with 20 ft-lb of torque for transporting purposes.[3] An appropriate torque applied once to the valve should give a good mechanical seal. Leakage might be detected by having a small line run from the valve cap to a detector. If it were anticipated that the valve might give too much trouble, the usual valve could be omitted and the cylinder could be filled initially with radioactive krypton through a welded-in-place fitting that could be welded shut. With the proper choice of materials and welding, this could give a more reliable seal than the usual valve. This seal would still allow retrievability although not as conveniently as with the foregoing valve.

Apart from the effects of rubidium and impurities, the cylinder and its closure, being entirely metallic, should have good radiation and thermal resistance. Another plus is the probable ease of recovery of the krypton-85 should it ever be required. The matter of cost has been estimated for krypton-85 stored at 500 psi in gas cylinders in the Reference Krypton Storage Facility.[11,17,18] This facility, sized for 2000 metric tons of fuel per year, would cost $208,500,000 (1976 dollars) over its 40 years of operation or about $5,210,000/year (1976 dollars) on the average. The cost probably could be reduced by using a higher gas pressure to reduce the number of bottles and size of the facility, but this would increase the risk of serious leakage.

On balance, the storage of krypton-85 in gas cylinders has often been considered to be one of the more favorable methods.[9,10,20-24] Two of the principal uncertainties are in the areas: (a) the possible sudden failure of a high-pressure cylinder or its valve, whether or not from corrosion or other causes; and (b) the effects of gaseous impurities in aggravating the corrosion of the cylinder and its closure.

23<

REFERENCES FOR GAS IN 500- OR 2000-PSI CYLINDERS

1. C.L. Bendixsen and F.O. German, "Operation of the ICPP Rare Gas Recovery Facility during Fiscal Year 1970," Report ICP-1001, Idaho Operations Office, Idaho Falls, Idaho, Oct 1971.

2. C.L. Bendixsen et al., "1972 Operations of the ICPP Rare Gas Recovery Facility," Report ICP-1023, Idaho Operations Office, Idaho Falls, Idaho, Mar 1973.

3. C.W. Nielsen, "Safety Analysis Report for Packaging Krypton Shipping Container USA/5958/BL (ERDA-ID)," Report ICP-1077, Idaho National Engineering Laboratory, Idaho Falls, Idaho, June 1975.

4. J.S. Davis and J.R. Martin, "A Cryogenic Approach to Fuel Reprocessing Gaseous Radwaste Treatment," Transactions of the American Nuclear Society, 1973 Annual Meeting, June 10-14, 1973, p 176, American Nuclear Society, Hinsdale, Ill. 60521.

5. H. Yusa et al., "Development of a Cryogenic Krypton Separation System for the Off-Gas of Reprocessing Plants," of Dr. M.W. First, ed., 15th DOE Nuclear Air Cleaning Conference, Boston, Mass., Aug 7-10, 1978, Harvard Air Cleaning Laboratory, 665 Huntington Ave., Boston, Mass. 02115.

7. J.R. Merriman et al., "Removal of Radioactive Krypton and Xenon from Contaminated Off-Gas Streams," of M.W. First and J.R. Morgan, Jr., eds., Proceedings of the Eleventh AEC Air Cleaning Conference, Richland, Wash., Aug 31-Sept 3, 1970, Harvard Air Cleaning Laboratory, 665 Huntington Ave., Boston, Mass. 02115, Dec 1970.

8. W.D. Burch et al., "Retention of Gaseous Fission Products in Reprocessing LMFBR Fuels," Report IAEA-AG-63-2, from advisory group meeting on reprocessing of Liquid Metal Fast Breeder Reactor Fuels, Leningrad, USSR, May 17-21, 1976, International Atomic Energy Agency, Vienna, Austria, May 1976.

9. B.A. Foster and D.T. Pence, "An Evaluation of High Pressure Steel Cylinders for Fission Product Noble Gas Storage," Report ICP-1044, Idaho Operations Office, Idaho Falls, Idaho, Feb 1975.

10. B.A. Foster et al., "Long Term Storage Techniques for ^{85}Kr," M.W. First, ed., Report CONF-740807-Pl, p 293, Proceedings of the Thirteenth AEC Air Cleaning Conference, San Francisco, Calif., Aug 12-15, 1974.

11. AEC Manual, Chapter 0529 "Safety Standards for the Packaging of Radioactive and Fissile Materials."

12. Code of Federal Regulations, Vol. 49, Transportation, Part 173, Oct 1, 1977.

13. R.A. Brown et al., "Reference Facility Description for the Recovery of Iodine, Carbon and Krypton from Gaseous Wastes," Report ICP-1126, National Technical Information Service, U.S. Department of Commerce, Springfield, Va., Apr 1978.

14. T.R. Pinchback and D.A. Knecht, "The Development of Process and Storage Materials Suitable for Krypton-85 Waste Management," paper presented at the International Symposium on Science Underlying Radioactive Waste Management, Boston, Mass., Nov 29-Dec 1, 1978.

15. R.W. Benedict et al., in C.M. Slansky, ed., "Technical Division Quarterly Progress Report, Oct 1-Dec 31, 1977," Report ICP-1141, DOE Idaho Operations Office, Feb 1978.

16. R.W. Benedict et al., in C.M. Slansky, ed., "Technical Division Quarterly Progress Report, July 1-Sept 30, 1977," Report ICP-1132, DOE Idaho Operations Office, Oct 1977.

17. "Capital Cost Estimate and Construction Impacts, Volatile Radioisotope and Off-Gas Treatment. Reference Facility for Krypton Storage," Report ICP-1127 (to be published), Idaho National Engineering Laboratory, Idaho Falls, Idaho.

18. D.A. Knecht, "An Evaluation of Methods for Immobilizing Krypton-85," Report ICP-1125, Idaho National Engineering Laboratory, Idaho Falls, Idaho, July 1977.

19. C.L. Bendixsen and J.A. Buckham, "General Survey of Techniques for Separation and Containment of Noble Gases from Nuclear Facilities," Report CONF-730915-16, from Symposium on Noble Gases, Las Vegas, Nev., Sept 24-28, 1973, Allied Chemical Corporation, Idaho Falls, Idaho, 1973.

20. C.L. Bendixsen and D. Knecht, "Separation and Storage of Krypton," p 343 in Proceedings of the International Symposium on the Management of Wastes from the LWR Fuel Cydle," Denver, Colo., July 11-16, 1976, Report CONF 760701, National Technical Information Service, U.S. Department of Commerce, Springfield, Va., 1976.

21. J.O. Blomeke and J.J. Perona, "Management of Noble-Gas Fission-Product Wastes from Reprocessing Spent Fuels", Report ORNL-TM-2677, Oak Ridge National Laboratory, Oak Ridge, Tenn., Nov 21, 1969.

22. R.A. Brown et al., Section 14.0, "Immobilization of Volatile Radioisotopes", of J.W. Bartlett et al., eds., "Alternatives for Managing Wastes from Reactors and Post Fission Operations in the LWR Fuel Cycle", Report ERDA-76-43, Vol 2, National Technical Information Service, U.S. Department of Commerce, Springfield, Va., May 1976.

23. J.P. Nichols and F.T. Binford, "Status of Noble Gas Removal and Disposal", Report ORNL-TM-3515, Oak Ridge National Laboratory, Oak Ridge, Tenn., Aug 1971.

24. R.P. Randl, "Waste Management in the Federal Republic of Germany", p 157 in Proceedings of the International Conference on Nuclear Power and its Fuel Cycle, Salzberg, Austria, May 2-13, 1977, Vol 4, International Atomic Energy Agency, Vienna, Austria, 1977.

2.2 ZEOLITES

Zeolites are framework silicates of aluminum, silicon, and oxygen with sodium, calcium and potassium as cations.[1,2] The cage unit within the silicate framework of some zeolites will accommodate the krypton atom. Emplacement into the cage must be done at high temperatures and high pressures. Once krypton is emplaced, the release rate from the cage is very slow at storage temperatures and pressures.[3,4]

References listed at the end of this section are concerned with the question of technical and, to a limited extent, economic feasibility of loading radioactive krypton on zeolites, primarily sodalite. Allied Chemical Corporation at the Idaho Chemical Processing Plant (ICPP) has studied extensively the requirements of operating a sorption unit and of the capacity of sodalite.[5,6]

Figure 2.2.1 is a schematic of the ICPP process of encapsulation. Since the process of sorption depends on the operating temperature and pressure as well as on the microscopic properties of the zeolite, (such as determined by the manufacturing process) additional information is needed to optimize the process with respect to pressure, temperature, type of sorbent, and time for sorption to occur. The effect of xenon is unknown. However, the data developed at the ICPP allow a preliminary evaluation of the technical feasibility and economics for sorption of krypton-85 on zeolites.[5]

Krypton sorption onto the zeolite must be accomplished at high temperature and high pressure (nominally 500° C and 7500 psi). Sorption does not occur instantaneously. Data from Knecht[5] show that a 25-cm^3 Kr/g loading (at 450° C) could be obtained at 180 MPa (26,000 psi) within 2.5 hr, 140 MPa (20,300 psi) within 4 hr, and 28 MPa (4060 psi) under equilibrium conditions.

The ICPP current work on the use of porous-metal-walled tubes for packaging krypton-85 has shown a unique advantage in that the need for handling krypton-loaded zeolite powders is eliminated. A package of porous metal tubes could be placed in larger containers for disposal. Thus, using porous metal tubes or containers for loading and disposal of the zeolites eliminates a handling problem which must be addressed in the other methods. That problem is the loading of krypton-85 onto the zeolite as powder or pills in the high-pressure vessel necessitates a solution to the problem of how to immobilize the loaded zeolite. A review of these immobilization methods has been prepared by Benedict[7] and is discussed in the following paragraph.

If loaded zeolites are stored as powder or pills placed in cylinders, the heat-transfer characteristics of the self-heating material in the cylinders must be determined. Christensen has

27<

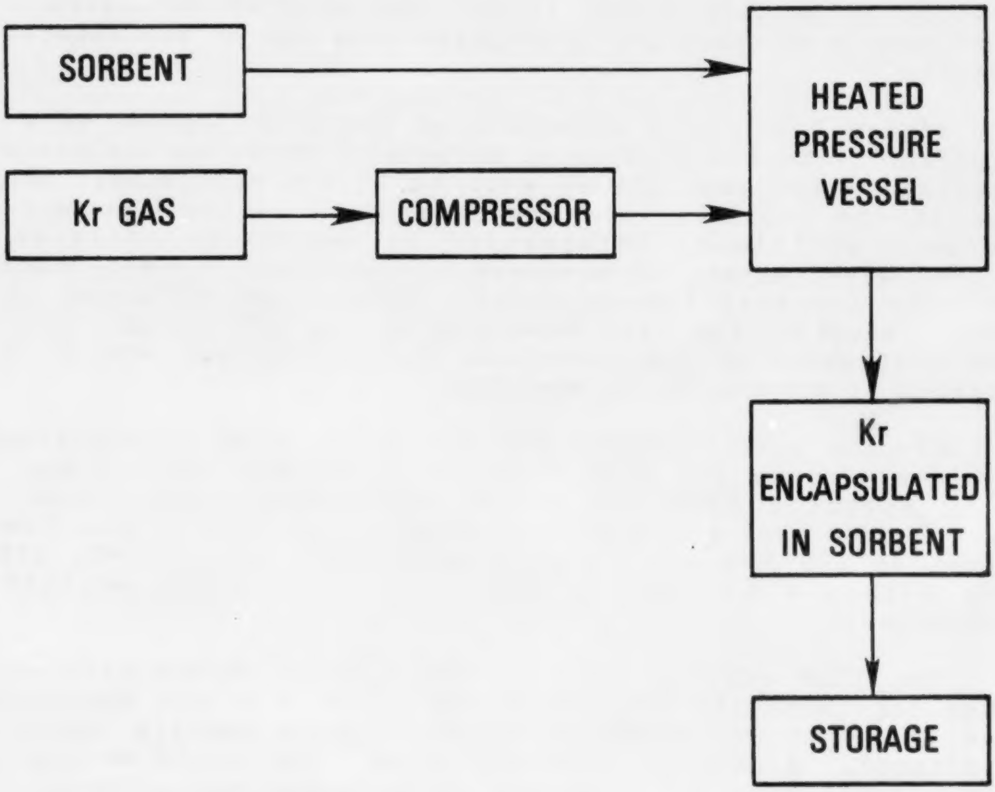

SORBENT
Kr GAS
COMPRESSOR
HEATED PRESSURE VESSEL
Kr ENCAPSULATED IN SORBENT
STORAGE

FIGURE 2.2.1. PROCESS FOR LOADING KRYPTON INTO SORBENT.

28<

performed some of the necessary analysis.[8] Refinements to his analysis could define more clearly the probability and consequences of release of krypton-85 from the matrix, and the possible consequences of the self-heating and lower heat transfer through the porous bed packed into the cylinders. D. A. Knecht[5] has taken the maximum loading on sodalite to be about 26 cm^3 of krypton (STP)/cm^3 of bulk zeolite. With this the sodalite in a 23-cm diameter cylinder would have a maximum temperature of 150° C and the leaked krypton would be less than 1% at any time. Zeolite samples loaded with krypton have been exposed at the Idaho National Engineering Laboratory (INEL) to gamma radiation for 28 days at temperatures of between 52°-68° C.[10] No leakage was observed in oven-dried, leached sodalite, but more than 80% leakage was observed in zeolite A. Another test will be described in a forthcoming technical-division progress report from the INEL. An evacuated tube filled with krypton-loaded sodalite was irradiated in a fuel storage facility at about 10^6-gamma rads/hr for 105 days. The temperature averaged 60° C with an occasional excursion to 115° C due to the imperfect control of conditions. At the end of the test neither the irradiated sample nor a control showed as much as 1% leakage of krypton. A more controlled experiment also is planned. Ultimately, tests also should be made with the appropriate exposure to beta radiation.

The waste management programs in Canada,[11] Germany,[12] and Japan,[13] also have examined zeolites for krypton disposal.

The review of the literature on loading zeolites with krypton indicates:

(a) Compact packaging can be achieved by loading krypton onto zeolites.

(b) Technical feasibility of sorbing stable krypton on zeolites has been demonstrated; sorbing radioactive krypton on zeolites and the storage behavior have yet to be investigated completely.

(c) Economic considerations of the zeolite loading method have yet to be determined because the bench-scale process must be studied further.[9]

REFERENCES FOR ZEOLITES

1. W.A. Deer, R.A. Howie, and J. Zussman, "An Introduction to Rock-Forming Materials", p 375, Longman Group Limited, London, 9th impression, 1976.

2. D.W. Breck, "Zeolite Molecular Sieves: Structure, Chemistry and Use" John Wiley & Sons, New York, N.Y. 1974.

3. R.M. Barrer and J.R. Cole, "Interaction of Sodium Vapour with Synthetic Sodalite: Sorption and Formation of Colour Centres" Journal of Physical Chemistry Solids 29, pp 1755-1758.

4. R.M. Barrer and D.E.W. Vaughan, "Trapping of Inert Gasses in Sodalite and Cancrinite Crystals", Journal of Physical Chemistry Solids 32, pp 731-743.

5. Dieter A. Knecht, "An Evaluation of Methods for Immobilizing Krypton-85", Report ICP-1125, Idaho National Engineering Laboratory, July 1977.

6. R.A. Brown, M. Hoza, and D.A. Knecht, "^{85}Krypton Storage by Zeolite Encapsulation", 14th ERDA Air Cleaning Conference, Allied Chemical Corporation, Idaho Chemical Programs, Idaho National Engineering Laboratory, Idaho Falls, Idaho.

7. Robert W. Benedict, "An Evaluation of Methods for Immobilizing Solids Loaded With Krypton-85", Report ICP-1130, Allied Chemical Corporation, Idaho Chemical Programs, Aug 1977.

8. A.B. Christensen, "Physical Properties and Heat Transfer Characteristics of Materials for Krypton-85 Storage", Report ICP-1128, Allied Chemical Corporation, Idaho Falls, Idaho, Sept 1977.

9. T.R. Pinchback and D.A. Knecht, "The Development of Process and Storage Materials Suitable for Krypton-85 Waste Management", paper presented at International Symposium on Science Underlying Radioactive Waste Management, Boston, Mass., Nov 29-Dec 1, 1978.

10. R.W. Benedict et al., "Krypton-85 Storage Development", pp 11-21 of C.M. Slansky, et al., "Technical Division Quarterly Progress Report, July 1-Sept 30, 1977", Report ICP-1132, Idaho National Engineering Laboratory, Idaho Falls, Idaho Oct 1977.

11. M. Tomlinson, S.A. Mayman, H.Y. Tunmemagi, W.F. Merritt, J.A. Morrison, H.S. Iruine, G.A. Vivina, J. Gale, B. Sanford, and P.J. Dyne, "Management of Radioactive Wastes from Nuclear Fuels and Power Plants in Canada," Report IAEA-CN-36/178.

12. R.D. Penzhorn, "Alternative Processes for the Long-Term Storage of KR-85," Report KFK-2482, Kernforschungszentrum Karlsruhe, G.m.b.H., Germany, F.R. (in German), Dec 1977; Atomindex 9, 396106 (1978).

13. Bunzai An, Kobe Steel Co., Wakinohama Ch 1-1, FUKIAI-ku, Kobe, Japan.

2.3 KRYPTON-LOADED SOLID SET IN A MATRIX

Krypton absorbed into, or adsorbed onto, solids such as zeolites, metals, charcoal or glass immobilizes the krypton, but the solid loaded with krypton must be stored. Several methods for handling these solids have been suggested by Benedict: encapsulation in bitumen, hydraulic cement, pressed (isostatic) blocks, metal matrix, glass matrix, and organic polymer matrices.[1] Table 2.3.1 contains a list of the processes considered, together with updated information. Factors which limit the utility of each process include radiation damage to the continuous media and the high temperature required for glass matrix formation.

Bitumen and polymer as continuous media are particularly susceptible to radiation damage. To prevent problems due to radiation damage, the loading of krypton must be kept low. This leads to an appreciable volume increase (factor of 10) over the original material containing the krypton. The storage space required and the handling problems consequently are increased. Radiation damage to the matrix could result in accelerated leakage which could become unacceptable.

Glass matrices would require such high temperature during formation that the krypton would leak from the solid. Suitable low-melting point alloys could eliminate this problem for cast metal matrices.[2] Examples of possible metals and alloys are given in Table 2.3.2. The melting points of these alloys are low enough that leakage of krypton from the zeolite during matrix formation would not be problem. This technique would be quicker than sintering with a metal powder, and probably easier than pressing with a metal powder. The metal could be allowed to flow into the porous metal tube after the zeolite had been loaded with krypton and evacuated. Metal matrices would lower the maximum temperature of the zeolite and thus reduce krypton leakage from it.

It might be possible to improve sufficiently the thermal technology to deposit on them a nickel coating of the desired thickness. This might be done by passing gaseous nickel carbonyl $Ni(Co)_4$ into the zeolite, the deposition temperature being in the range of 100°-200° C. The gaseous carbonyl could be made easily and economically at the krypton-loading area. Nickel carbonyl is a poisonous gas, but it might not be any more dangerous to handle than gaseous radioactivity.

The alternatives which seem favorable are porous metal containers, nickel-coated zeolite, and possibly metal matrices of cast metal or pressed powder. Table 2.3.3 is a summary of the advantages and disadvantages of using matrices to immobilize the krypton-loaded solids. The effect of the daughter rubidium usually would not be serious.

TABLE 2.3.1

COMPARISON OF CANDIDATE PROCESSES FOR SOLID STORAGE

Process	Process Temp oC	Maximum ^{85}Kr Loading[a] (cm3(STP)/cm3 solid)	Limiting Factor
Porous Metal Container	-	26[c]	Coordination with gas immobilization equipment
Matrix of Pressed Metal Powder	250-500	26[c]	Complicated processing equipment, cost
Matrix of Cast Metal	200-300	26[c]	M. pt. of alloy, cost
Nickel Coating on Zeolite	100-200	26[c]	Control of deposition, hazardous gas
Matrix of Sintered Metal	250-700	26[c]	Cost, process temperature
Matrix of Polymer	100	2.6[d]	Radiation damage to polymer
Matrix of Cement	25	0.06[d]	Radiation damage to cement[b]
Matrix of Bitumen	150-230	2.6[d]	Radiation damage to bitumen; storage temperature could not exceed 80° C
Matrix of Glass	900-1070	-	High process temperature

---

[a] Loading possible without radiation damage to matrix or loss of krypton from sodalite

[b] The effect of water is not large; see Hydrofracture

[c] See reference 3

[d] See reference 1

TABLE 2.3.2

SOME METALS AND ALLOYS FOR A POSSIBLE MATRIX

Composition (wt%)	M. Pt. (°C)	Density (g/cm^3)	Cost (¢/cm^3 matrix)[4]
63 Sn + 37 Pb	183	7.72	7.0
91 Sn + 9 Zn	198	6.55	9.2
100 Sn	232	6.5	9.1
100 Bi	271	9.8	6.5
100 Pb	328	11.3	0.85
(100 Ni	1453	8.90	4.1)

34<

TABLE 2.3.3

ADVANTAGES AND DISADVANTAGES OF KRYPTON LOADED SOLID SET IN A MATRIX

Advantages	Disadvantages
The volume of matrix for disposal can be kept relatively small for some matrix materials.	Some nonmetallic binders are damaged by radiation which likely results in leakage.
Good heat transfer from the matrix and resistance to radiation are possible with metal matrices, and high loadings of the krypton-loaded solid can be achieved. Resultant package of waste is compact.	To decrease radiation damage to nonmetallic binders, loading of krypton - containing solid must be low, with a resultant increase in volume of matrix to be disposed of.
Some secondary containment and gamma shielding are provided by the matrix.	Increased expense of production per unit volume of zeolite.
Solids (zeolite pills, for example) loaded with krypton would be easier to handle than charcoal.	

REFERENCES FOR KRYPTON LOADED SOLIDS SET IN A MATRIX

1. Robert W. Benedict, "An Evaluation of Methods for Immobilizing Solids Loaded with Krypton-85", Report ICP-1130, Allied Chemical Corporation, Idaho Chemical Programs, Aug 1977.

2. K.M. Lamb, "Final Report: Development of a Metal Matrix for Incorporating High-Leval Commercial Waste", Report ICP-1144, Allied Chemical Corporation, Idaho Chemical Programs, Mar 1978.

3. D.A. Knecht, "An Evaluation of Methods for Immobilizing Krypton-85," Report ICP-1125, National Technical Information Service, U.S. Department of Commerce, Springfield, Va., July 1977.

4. Metals Week, "Monthly Prices", Sept 1978, Averages, Oct 9, 1978.

36<

2.4 ION IMPLANTATION/SPUTTERING

This technique has been under development for several years;[1-9] its useful application to the entrapment of krypton-85 is currently being pursued at the Battelle Pacific Northwest Laboratories[10-18] and in New York,[19-20] Canada,[21-26] Europe[27] and Japan.[28] It has the advantage over certain other contenders of requiring no undesirable pressure or temperature either during the initial emplacement or during long-term storage.

The elements of the process for a crystalline host metal may be illustrated by reference to the cylindrical arrangement of Figure 2.4.1.[2,10,29] The krypton is admitted to the evacuated apparatus at 0.1-100 Pa and ionized to Kr^+. The Kr^+ cation is attracted continuously to the target which is covered with the material that is to serve as the host for the krypton. However, this material is sputtered to the cooled substrate where it also receives Kr^+ ions on the negative part of the indicated half-wave. The lower average voltage on the substrate compared with the target allows a net build-up of sputtered, krypton-implanted host material on the cylindrical substrate from which it may be later separated. The process proceeds near room temperature. The half-wave rectified waveform tends to produce a stable plasma. When arcing does occur, however, there still is a need for a fast-acting, high-current switch. The interference from the contaminant xenon apparently is not serious according to a communication of Dr. G. L. Tingey, Battelle Pacific Northwest Laboratories.

The host material is usually a metal such as nickel (Figure 2.4.1), aluminum, iron, titanium, or an alloy. For example, a 10.4-liter (STP) quantity of krypton was so implanted in 744 g of nickel to give 14 cm^3 (STP) Kr/g Ni, or 124 cm^3 (STP) Kr/cm^3 Ni in a 3-mm thickness.[10] The temperature for a given fractional release of implanted krypton increases with the recrystallization temperature of the metal. This temperature is thus higher for nickel (600° C) than it is for aluminum (300° C). Oxidation or other changes at the surface of these "kryptonates" would release krypton; hence, a cap or close-out layer of the same metal without krypton is appropriate before handling. It is found that sputtered alloys in the amorphous or glassy state can contain markedly higher loadings of krypton than do the crystalline metals.[20] Thus, amorphous 90% iron-10% yttrium has been loaded to 8.2 atomic % krypton, which amounts to about 178 cm^3 (STP) Kr/cm^3 alloy.[17] Still higher loadings of over 400 cm^3 (STP) Kr/cm^3 alloy may be estimated from data on thin amorphous films of $Gd_{0.11}Co_{0.73}Mo_{0.16}$ by Cuomo and Gambino.[20,27] The above loadings may be compared with 100 cm^3 (STP) Kr/cm^3 of space for gas storage at 2000 psi.

The heat-transfer properties of crystalline metals pose no problem. A thermal conductivity of 0.01 w $cm^{-1}K^{-1}$

37<

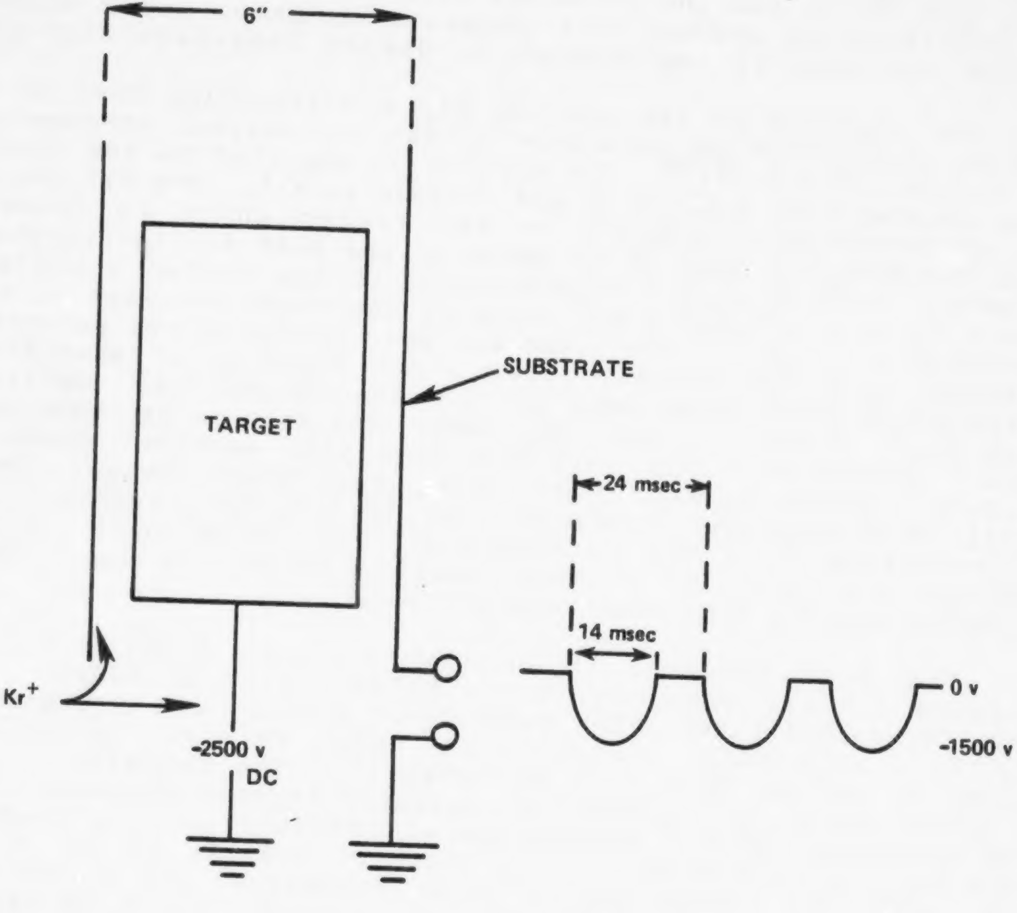

6"
SUBSTRATE
TARGET
24 msec
14 msec
0 v
-1500 v
Kr$^+$
-2500 v
DC

FIGURE 2.4.1 EXPERIMENTAL DIAGRAM FOR ION IMPLANTATION/SPUTTERING

in an amorphous alloy such as $Gd_{0.11}Co_{0.73}Mo_{0.16}$ would not restrict its loading below 400 cm^3 (STP) Kr/cm^3 alloy in order to avoid the recrystallization of the alloy and rapid release of krypton at 800° C.[29] This appears not to be a practical limitation.

The power requirements to handle the krypton-85 from 2000 metric tons of fuel per year would not be large at the 100 cm^3 (STP) Kr/cm^3 metal that is readily attainable by ion implantation/sputtering.[17,29,30] The prospect for scaling up the equipment appears to be good if amorphous alloys are used, thus allowing only a direct current substrate voltage and simpler operation.[17] The apparatus recently has been increased in size about 100% over that of Figure 2.4.1.

The technique requires further work in the control of arcing, the uniformity of deposits,[10] the effects of rubidium, radiation, corrosion and diffusion, for example, but no insurmountable problems appear to be present. The high product density should lead to less expensive storage, and the product should be readily retrievable. The heat transfer properties are probably better than those of zeolites.[29] A small amount of the loaded krypton can be expelled in several hours at low temperature. The remainder in an uncapped, amorphous iron-yttrium alloy has been shown to be released with an Arrhenius-type dependence on temperature.[17] Measurements above 350° C have suggested that the maximum leaked amount at any time at 200° C would never exceed 0.1% of the original loading for this material. An iron-zirconium alloy is believed to leak an order of magnitude less than the iron-yttrium alloy.[17] Hence early measurements on leakage do not indicate a serious problem.

REFERENCES FOR ION IMPLANTATION/SPUTTERING

1. K.B. Blodgett and T.A. Vanderslice, "Mechanisms of Inert Gas Cleanup in a Gaseous Discharge," Journal of Applied Physics 31, 1017 (1960).

2. G. Carter and J.S. Colligan, "Ion Bombardment of Solids," American Elsevier, New York, N.Y., pp 354-422, 1968.

3. R.S. Nelson et al., "Improvements in or Relating to the Storage of Material," British Patent 1,485,266/A/ to UKAEA, Sept 8, 1977.

4. A.D. LeClaire and A.H. Rowe, Report A.E.R.E. M/R 1414, p 1, Atomic Energy Research Establishment, Harwell, England, 1957.

5. G. Carter et al., "Ion Sorption in the Presence of Sputtering," Procedures of Physics Society 79, 299 (1962).

6. O. Almen and G. Bruce, Nuclear Instrumentation Methods 11, 257 (1961).

7. D. Chleck et al., "Radioactive Kryptonates - I. Preparation," International Journal of Applied Radiation and Isotopes 14, 581 (1963).

8. W.A. Grant and J.N. Baruah, "Ion Implantation Damage in GaAs Measured by the Channeling of ^{85}Kr," Radiation Effects 17 (1-2), 13-17 (Jan 1973).

9. D. Chleck et al., "Radioactive Kryptonates - II Properties," International Journal of Applied Radiation and Isotopes 14, 593 (1963).

10. M.A. Bayne et al., "Krypton Entrapment in Pulse-Biased Sputter-Deposited Metals," Report PNL-SA-6698, presented at the Third International Conference on Metallurgical Coatings sponsored by American Vacuum Society in San Francisco, Calif., Apr 3-7, 1978, Battelle Pacific Northwest Laboratories, Richland, Wash.

11. E.D. McClanahan et al., "State of the Art for High Rate Sputter Deposition," presented at Workshop on Alternatives for Cadmium Electroplating in Metal Finishing, National Bureau of Standards, Gaithersburg, Md., Oct 4-5, 1977.

12. G.L. Tingey, Sect 3 of A.H. Platt (comp.), "Nuclear Waste Management and Transportation Quarterly Progress Report, July Through September, 1976," Report BNWL-2179, Battelle Pacific Northwest Laboratories, Richland, Wash., Jan 1977.

13. E.D. McLanahan et al., "Krypton Entrapment During Sputtering," pp 4.2-4.6 of A.M. Platt (comp.), Nuclear Waste Management Quarterly Progress Report, January through March 1977, Report BNWL-2290, National Technical Information Service, U.S. Department of Commerce, Springfield, Va., July 1977.

14. E.D. McLanahan et al., "Krypton Entrapment During Sputtering," of A.M. Platt (comp.), "Nuclear Waste Management Quarterly Progress Reports, April through December 1977," Reports BNWL-2377/2 and PNL-2377-3 and-4, National Technical Information Service, U.S. Department of Commerce, Springfield, Va., May 1978.

15. M.A. Bayne et al., "Krypton Entrapment During Sputtering," of A,M, Platt and J.A. Powell (comp.), "Nuclear Waste Management Quarterly Progress Report, January through March 1978," Report PNL-2378-1, National Technical Information Service, U.S. Department of Commerce, Springfield, Va., Aug 1978.

16. G.L. Tingey, in "Nuclear Waste Management Quarterly Progress Report for April-June 1978," Report PNL-2378-2, National Technical Information Service, U.S. Department of Commerce, Springfield, Va., Nov 1978.

17. G.L. Tingey et al., "Entrapment of Krypton in Sputter Deposited Metals - A Storage Medium for Radioactive Gases," Report PNL-2879, Pacific Northwest Laboratory, Richland, Washington 99352, Jan 1979.

18. M.A. Bayne et al., "Krypton Entrapment in Pulse-Biased Sputter-Deposited Metals," Thin Solid Films, 54, 327 (1978).

19. J.J. Cuomo and R.J. Gambino, Journal of Vacuum Science Technology 12 (7), 79 (1975).

20. J.J. Cuomo and R.J. Gambino, "Incorporation of Rare Gases in Sputtered Amorphous Metal," Journal of Vacuum Science Technology 14 (1), 152 (1977).

21. F. Brown, Report of Solid-State Science Branch, pp 1-2, "Chemistry and Materials Division Progress Report, July 1, 1973-September 30, 1973," Report AECL-4671, Atomic Energy of Canada Ltd., Chalk River, Ontario, Nov 1973.

22. J.A. Davies, J. Friesen and J.D. McIntyre, Canada Journal of Chemistry 38, 1526 (1960).

23. J.A. Davies et al., Canada Journal of Chemistry 38, 1535 (1960).

24. J.A. Davies and G.A. Sims, Canada Journal of Chemistry 39, 601 (1961).

25. J.A. Davies et al., Canada Journal of Chemistry 39, 611 (1961).

26. J.A. Davies et al., Arkiv. F. Fysik. 24, 377 (1963).

27. H.J. Matzke, "Krypton Release from Nickel and Stainless Steel," Vacuum 25 (9/10), 407 (1975).

28. S. Kitani, "Application of Ion Implantation Technique To Fixation of Kr-85," pp 63-67, Report JAERI-M-5567, from meeting on atomic energy research with heavy ions, Tokai, Ibaraki, Japan, Oct 25, 1973, (in Japanese); NSA 31, 33361 (1975).

29. D.A. Knecht, "An Evaluation of Methods for Immobilizing Krypton-85," Report ICP-1125, Allied Chemical Corporation, Idaho Falls, Idaho, July 1977.

30. J.W.S. Bodys and K.C. Campbell, "Mechanism of the Kryptonation of Nickel by Film Evaporation," Journal of Chemistry Society, Faraday I, 70, 2170-2179 (1974).

BIBLIOGRAPHY FOR ION IMPLANTATION/SPUTTERING

R.S. Berg et al., Journal of Vacuum Science Technology 11, 52 (1974).

S.L. Blum and J. Pappis, Electronic Progress 4, (6), 1 (1960).

K.L. Chopra, Thin Film Phenomena, p 200, McGraw-Hill Book Co., New York, N.Y.

G. Dearnaley et al., "Range Distribution of Radioactive Ions Implanted into Silicon Crystals," pp 635-656 of D.W. Palmer, ed., Atomic Collision Phenomena in Solids, North Holland Publishing Co., London, 1970.

F.H. Eisen and B. Welch, European Conference on Ion Implantation, p 227, Peter Peregrinus Ltd., England, 1970.

E.A. Fagen et al., Journal of Applied Physics 45, 50 (1974).

G. Helm and E. Kay, "Ion Implantation during Film Growth and Its Effect on the Superconducting Properties of Niobium," Journal of Applied Physics 46, 4006 (1957).

F.M. D'Heurle, Metall. Transactions 1, 725 (1970).

R. Kelly and E. Ruedl, "The Diffusion in Inert-Gas Bombarded Pt and Al," Phys. Stat. Sol. 13, 55 (1966).

A.D. LeClaire and A.H. Rowe, "The Diffusion of Argon in Silver," Rev. Met. 52, 94 (1955).

W.W.Y. Lee and D.W. Oblas, Journal of Applied Physics 46, 1728 (1975).

W.W.Y. Lee and D.W. Oblas, Journal of Vacuum Science Technology 7, 129.

P. Lukac, "Sectioning Technique for SiO_2 and Its Application to Penetration Studies," Radiochem Radioanal. Lett. 18 (4), 203-209 (Aug 21, 1974).

I.V. Mitchell and R.C. Madison, "Gas Incorporation in Sputtered and Evaporated Gold Films," Vacuum 21, 591.

J.W. Mayer et al., "Ion Implantation in Semiconductors Silicon and Germanium," Academic Press, New York, N.Y., 1970.

L.I Maissel, Handbook of Thin Films Technology, pp 4-25, McGraw-Hill Book Co., New York, N.Y., 1970.

D.M. Mattox and G.J. Kominiak, Journal of Vacuum Science Technology 8, 194 (1971).

D.E. Polk, Acta Metall. 20, 485 (1972).

D.E. Rimmer and A.H. Cottrell, Phil. Mag. 8, 1345 (1957).

D. Rosenberg and G.K. Wehner, Journal of Applied Physics, V, 33, (5) 1842 (1962).

G.G. Schwartz and R.E. Jones, IBM J. Res. Div. 14, 52 (170).

D.C. Santry, "Study of Dose Effects on the Retention of Ions Implanted at keV Energies," pp 300-306, Report CONF-730618, from 8th International EMIS Conference on Low Energy Ion Accelerators and Mass Separators held in Skovde, Sweden, June 12, 173 Chalk River Nuclear Laboratories, Ontario, Canada, Aug 1973.

S. Thulin, Ark. Fys. 9, (10), 107 (1955).

J.M. Tobin, "The Diffusion of Krypton in Silver," Acta Metal 5, 398 (1957).

J.L. Vossen, Journal of Vacuum Technology 8, 812 (1971).

H.F. Winters and E. Kay, "Gas Incorporation Into Sputtered Films," Journal of Applied Physics 38, 3928 (1967).

J.L. Whitton et al., Journal of Materials Science 4, 208 (1969).

H.F. Winters and E. Kay, Journal of Applied Physics 43, 794 (1972).

W.H. Weisenberger et al., Radiation Effects 9, 121 (1971).

2.5 METAL VAPOR DEPOSITION

Krypton at 0.1-100 Pa becomes uniformly absorbed in a film of metal condensing at liquid-nitrogen temperatures as has been shown for nickel, copper and iron.[1] There is no applied electric field. The condensing metal may have been vaporized previously by any means. Krypton uptakes of up to 6 atom % Kr have been obtained in nickel condensed at the above -196° C, but no uptake has been observed at condensing temperatures of 0° C or -78° C.[2] If the film containing 6 atom % Kr is warmed to room temperature, all but 0.23 atom % Kr (1 cm^3 (STP)/g Ni) is released. The release continues at a lower rate at higher temperatures. The highest loadings have been obtained with nickel. The work has not been extended beyond thin films. The heat-transfer characteristics of the film may be similar to those from ion implantation.[3]

Although the power requirements[4] and cost of nickel for this process appear moderate, further work probably would be necessary to improve the krypton-loading and possibly to retard the leakage. This technique has been used to prepare "kryptonates" to serve as indicators in analytical chemistry and other areas.[5] Any attack of the surface metal liberates krypton, an undesirable characteristic for the storage of krypton-85.[6]

REFERENCES FOR METAL VAPOR DEPOSITION

1. J.W.S. Bodys and K.C. Campbell, "Radioactive Kryptonation of Metals by Film Evaporation," International Journal of Applied Radiation and Isotopes 24, 107 (1973).

2. J.W.S. Bodys and K.C. Campbell, "Mechanism of the Kryptonation of Nickel by Film Evaporation," Journal of Chemical Society, Faraday Transactions 1 (12), 2170 (1974).

3. A.B. Christensen, "Physical Properties and Heat Transfer Characteristics of Materials for Krypton-85 Storage," Report ICP-1128, National Technical Information Service, U.S. Department of Commerce, Springfield, Va., 22161, Sept 1977.

4. D.A. Knecht, "An Evaluation of Methods for Immobilizing Krypton-85," Report ICP-1125, National Technical Information Service, U.S. Department of Commerce, Springfield, Va., 22161, July 1977.

5. J.E. Carden, "Radio-Release in Review with Special Emphasis on ^{85}Kr Clathrates and Kryptonates," Report ORNL-IIC-18, Oak Ridge National Laboratory, Oak Ridge, Tenn., July 1969.

6. D. Chleck and R. Maehl, "Radioactive Kryptonates - II Properties," International Journal of Applied Radiation and Isotopes 14, 593 (1953).

BIBLIOGRAPHY FOR METAL VAPOR DEPOSITION

1. A. C. Schaffhauser, ed., Proceedings of the Conference on Chemical Vapor Deposition of Refractory Metals, Alloys and Compounds, Gatlinburg, Tenn., American Nuclear Society, Hinsdale, Ill., Sept 12-14, 1967.

47<

2.6 CLATHRATES

H.M. Powell discovered that a krypton atom may be retained at room temperature in a cage of quinol molecules.[1] There are a number of variations on the preparation of the clathrate, but the techniques are generally easy.[2,3] A concentration of up to 61 cm^3 (STP) krypton/g has been achieved. Quinol appears to have the best properties for trapping krypton.[4] The quinol is stabilized in the clathrate at room temperature in the otherwise unstable beta form to which it gradually transforms with a loss of krypton. It is interesting that radiation seems to retard this transformation at room temperature and thereby slows the release of krypton. Cross-linking from the radiation appears to stabilize the beta form.[5] Contained krypton-85 does not irradiate the clathrate quickly enough to prevent its own leakage; therefore, irradiation of the clathrate immediately after its formation would probably be required to prevent excessive leakage. This retardation of the beta-alpha transition also has been observed when the beta quinol cages were empty.[6] Lindquist and Diethorn have suggested that sublimation controls the release of gas above room temperature.[5]

Although the clathrate is generally noncorrosive toward containers, the clathrate is itself subject to attack with a consequent loss of krypton. Most obviously, the clathrate is soluble in water and other solvents with an attendant loss of krypton. Hence, storage in a moisture-free atmosphere is considered to be prudent.[7] Certain gaseous oxidants such as ozone, sulfur dioxide, chlorine and flourine attack the clathrate.[8,9] If the clathrate containing the krypton-85 is exposed to air, the oxygen near the material is converted to ozone by radiolysis, and the latter oxidizes part of the quinol cage, releasing krypton-85. It can be represented by the chemical equation:[3]

$$[C_6H_4(OH)_2]_3[^{85}Kr] + O_3 = 3\,C_6H_4O_2 + 3\,H_2O + \text{free}\ ^{85}Kr$$

(This reaction without the prior radiolysis has been used to measure ozone at various elevations in the atmosphere.)[10,11]

The metallic rubidium daughter of krypton-85 probably would degrade the quinol clathrate; also, the rubidium oxide formed from the attack on the clathrate or from any presence of air would affect the clathrate.

The worst limitation of clathrates for storing krypton appears to be their thermal instability.[2] The effect of this instability is compounded by the poor thermal conductivity of organic compounds and the heating effect of krypton-85. The quinol clathrate begins to lose krypton at about 70° C and complete loss of krypton from the clathrate has been observed at 120° C, well below the 170° C melting point of quinol.[12,13] It is estimated by Foster et al. that the decay heat from the krypton-85 limits the clathrate loading to about 5 cm^3 (STP) Kr/g.[12] The reprocessing of 2000 metric tons of fuel would require 10^5 lb of quinol, an exorbitant amount.

The characteristics of quinol clathrates are summarized in Table 2.6.1.

49<

TABLE 2.6.1

ADVANTAGES AND DISADVANTAGES OF QUINOL CLATHRATES

Advantages	Disadvantages
Ease of formation.	Thermally unstable at useful krypton loadings because of poor thermal conductivity.
Believed to be non-corrosive.	High expense of quinol at 5 cm^3 (STP) Kr/g.
Apparent stability to radiation.	Susceptibility of attack of clathrate by oxygen, by the rubidium daughter of krypton, and by solvents such as water.
	A need to irradiate product immediately after formation to forestall excessive leakage.

REFERENCES FOR CLATHRATES

1. H.M. Powell, "The Structure of Molecular Compounds. Part VII. Compounds Formed by the Inert Gases," Journal of Chemical Society 1950, 298.
2. B.A. Staples and D.T. Pence, "An Evaluation of Quinol Clathrate for Fission Product Noble Gas Storage," Report ICP-1045, Allied Chemical Corporation, DOE Idaho Operations Office, Idaho Falls, Idaho, July 1974.
3. J.E. Carden, "Radio-Release in Review with Special Emphasis on ^{85}Kr Clathrate and Kryptonates," Report ORNL-IIC-18, Oak Ridge National Laboratory, Oak Ridge, Tenn, July 1969.
4. J.E. Mock et al., Industrial Engineering Chemistry 53, 1007 (1961).
5. K.O. Lindquist and W.S. Diethorn, International Journal of Applied Radiation and Isotopes 19, 333 (1968).
6. K.O. Lindquist and W.S. Diethorn, International Journal of Applied Radiation and Isotopes 20, 551 (1964).
7. E.J. Wilson and D.J. Hughes, International Journal of Applied Radiation and Isotopes 12, 129 (1961).
8. J.W. McLain and W.S. Diethorn, International Journal of Applied Radiation and Isotopes 15, 151 (1964).
9. D.J. Chleck and C.A. Ziegler, Nucleonics 17, 130 (1959).
10. D.J. Chleck et al., "Yearly Report on the Development of Radiochemical Analyzers for Non-Radioactive Gases and a Study of Radioactive Clathrates," Report AECU-4493, Tracerlab, Inc., 1959.
11. D.J. Chleck and C.A. Ziegler, "The Radiometric Analysis of Non-Radioactive Materials by Chemical Exchange," p 351 in Radioisotopes in the Physical Sciences and Industry, Vol II, Proceedings of Conference, Copenhagen, Denmark, Sept 6-17, 1960, IAEA, Vienna, Austria, 1962 (STI/PUB/20).
12. B.A. Foster et al., "Long Term Storage Techniques for ^{85}Kr," in Proceedings of 13th AEC Air Cleaning Conference, 1974, Report CONF-740807, National Technical Information Service, U.S. Department of Commerce, Springfield, Va., Vol 1, p 293, Mar 1975.
13. H.M. Powell, Journal of Chemical Society 1950, 300.

BIBLIOGRAPHY FOR CLATHRATES

1. H.G. Richter and A.S. Gillespie, Jr., "A Radio-Release Technique for the Measurement of Moisture in Gases," Final Report ORO-591, Research Triangle Institute, Jan 1, 1964.

2. Sister Martinette Hagan, "Clathrate Inclusion Compounds," Reinhold, New York, 1962.

3. J.E. Mock et al., "Crystallization of the Rare-Gas Cathrates," Industrial Engineering Chemistry 53, 1007 (1961).

4. D.J. Chleck and C.A. Ziegler, "The Preparation and Some Properties of Radioactive Quinol-Krypton Clathrate Compounds," International Journal of Applied Radiation and Isotopes 7, 141 (1959).

5. D. Chleck et al., "Detection Techniques for Hazardous Vapors of Elemental Propellants," Aeronautical Systems Division, Wright-Patterson Air Force Base, Report ASD-TR-63-294, Parametrics, Inc., Mar 1963.

6. R.J. Murgatroyd, "Ozone and Water Vapor in the Upper Troposphere and Lower Stratosphere," World Meteorology Organization, Technical Note No. 68, 68 (1965).

7. A. Tani, et al., "Method for Storing Radioactive Rare Gases," (In Japanese) Japanese patent document 1976-133697 /A/, May 15, 1975, Japan Patent Information Center, Tokyo; Atomindex 8 (19) 333130 (1977).

52<

2.7 POROUS UNDERGROUND MEDIA

One alternative for the ultimate disposal of krypton is emplacement underground in porous media. The literature on methods of gas injection is extensive, particularly the experience with injection of natural gas into underground storage areas to accommodate peak demand loads.[1,2,3] Specific literature on the subject of krypton disposal into underground porous media is available.[4-11] Site-specific information was developed during field tests conducted by INEL for xenon-133 disposal.[9] The depth of injection was 112 to 120 ft below the surface. The test demonstrated that a little (less than 1%) of the xenon-133 (half life 5.27 days) was lost to the atmosphere during the observation period which lasted about 5 half lives (26 days). For krypton-85, with a half life of 10.73 yr, this amount of leakage from shallow injection may not be tolerable. Whether this leak rate (less than 1%/month) would be a hazard would depend on the population exposed to the gas. In a remote location, such as the INEL or the Hanford reservation, it may be tolerable. Although the specific field tests were with xenon-133, it is reasonable to assume that krypton-85 would behave similarly since it is also a noble gas and would not undergo extensive sorption on the porous media. The chemical association in the soil solutions also would be unlikely. However, some of the gas does escape into the atmosphere. The general public may feel that even the very small doses from such leakage is unacceptable. Leaks in underground storage areas for natural gas have been managed to an acceptable degree.[4] A smaller scale field test using krypton-85 also was conducted at the INEL.[7] The injection depth was 10 to 15 ft. Transport through the soil was described by a heat flow-diffusion analog mathematical model. Acceptable agreement between the analysis and the model was found.

Underground disposal of krypton-85 has been addressed by Tadmor and Cowser.[5] These authors essentially conclude that underground disposal is technically feasible and provides acceptable protection to the public provided some specific geologic conditions exist at the disposal site. Molecular diffusion through the porous media must provide an adequate atmospheric release delay (about 100 yr) to be acceptable. However, locating an acceptable site where the necessary geologic conditions exist could be a difficult task. Also, testing to verify that the site is acceptable would require considerable field work.

Throughout the literature on storage underground in porous media, the criteria for an acceptable geologic site become clear.[1,3,12,13,14,15,16] The conditions are summarized in Table 2.7.1.

A significant collection of information on underground disposal exists in the thesis of P.C. Reist.[6] The informa-

TABLE 2.7.1

CRITERIA FOR POROUS MEDIA OF AN UNDERGROUND STORAGE SITE

A geologic zone of porous material of sufficient volume to hold the gas must exist.

Caprock structures of low permeability (10^{-16} to 10^{-18} cm^2) must exist over the storage zone to delay the migration to the ground surface-air interface for almost 150 yr.

The structure over the gas storage zone must be free of vertical cracks which might be a shorter path to the surface. Cracks leading horizontally from the storage zone are also undesirable, but less critical than the vertical cracks.

---

tion on the subject includes disposal costs and some of the basic data needed to evaluate a site (i.e., diffusivity of krypton through porous media). Although the cost data are dated, the relative cost advantage of underground storage in porous media over storage in pressurized cylinders should exist at today's costs. Reist[4] quotes costs (in 1964 dollars) for injection into underground media as about \$2/1000 ft^3 (STP) whereas other methods range from \$48 to \$227/1000 ft^3. The other primary method that was considered was storage in high-pressure cylinders in above-ground bunkers.

Cases of gas injection considered by Reist[4] and by Tadmor et al.[5] clearly show an economic advantage for underground disposal. Table 2.7.2 is a summary of the advantages and disadvantages of underground disposal. Major advantages of this alternative are the existence of experience with underground storage of gas, the ability to inject unrefined off-gas from the reprocessing plant, and the low cost of disposal of gaseous wastes. Also, the corrosion problems due to rubidium in steel containers are eliminated. After injection, the injection well could be plugged and only monitoring equipment left at the site.

Possibly the major disadvantage is public opinion about the safety of the injection operation and of the hazards of groundwater contamination. Experience with natural gas injection into the Herscher Dome near Chicago demonstrates that the leak rates are small, and that remedial action involving pumping out the leak zones and re-injecting the gas resulted in control of the storage site.[2] The impact of a small leak could be minimized simply by siting the injection well in areas remote from population centers and where the critera on Table 2.7.1 are satisfied.

A comprehensive review of the feasibility of underground storage has become available.[17] This study identifies possible advantages to this method, but also points to several areas of investigation which require additional study before underground storage could be undertaken.

In conclusion, underground storage in porous media appears to have a significant cost advantage over other alternatives. It appears that public acceptance of this disposal method would be the greatest barrier to its implementation. A reexamination of this alternative in view of advances in gas storage and oil drilling technology seems warranted.

TABLE 2.7.2

ADVANTAGES AND DISADVANTAGES OF POROUS UNDERGROUND MEDIA

Advantages	Disadvantages
Technology for injection of natural gas into porous media has already been demonstrated. Cost information, etc. are readily available.	May be difficult to assure that no leaks occur from the storage zone. Managing a slow leak is technically feasible, as demonstrated from natural gas storage experience.
No need to purify the off-gas to the same degree as in alternatives such as for storage in high pressure cylinders. Mixtures of gas, ^{133}Xe, ^{85}Kr, CO_2, H_2, O_2, N_2, could be injected without separation of ^{85}Kr, provided the storage zones had large enough capacity.	Locating and verifying that a site has the required geologic conditions may be difficult and costly. An acceptable site near a reprocessing plant may not be found, thus requiring expensive reprocessing and concentration of the gas for transportation to an acceptable site.
Cost of disposal is much lower than for above ground methods, approximately 1/25th the cost of the next lowest such alternative.	Public acceptance of this alternative may be difficult to gain.
No corrosion problems with the daughter rubidium in the porous media.	Gases injected into porous formation would not be as recoverable as would gases stored in high-pressure cylinders.

REFERENCES FOR DISPOSAL OF KRYPTON IN UNDERGROUND POROUS MEDIA

1. D.L. Katz, D. Cornell, R. Kobayashi, F.H. Poettman, J.A. Vary, J.R. Elenhaas and C.F. Weinaug, Handbook of Natural Gas Engineering, McGraw-Hill Book Co., New York, N.Y., 1959.

2. O.S. Hallden, "Underground Natural Gas Storage (Herscher Dome)", Proceedings of the 1961 Symposium on Ground Water Contamination, 110 R.A. Taft Sanitary Engineering Center, Cincinnati, Ohio, 1961.

3. R.J. Millington, "Gas Diffusion in Porous Media", Science 130, 100-102 (1959).

4. Parker C. Reist, "The Disposal of Waste Radioactive Gases in Porous Underground Media," Nuclear Applications 3, 474-480 (Aug 1967).

5. Tadmor, Jacob, and Cowser, "Underground Disposal of Krypton-85 from Nuclear Fuel Reprocessing Plants", Nuclear Engineering and Design, pp 243-250 (1967).

6. Parker C. Reist, "The Disposal of Radioactive Krypton-85 in Porous Media", Report NYO-841-7, Department of Industrial Hygiene School of Public Health, Harvard University, 665 Huntington Avenue, Boston, Mass. 02115, Oct 26, 1965.

7. John B. Robertson, "Diffusion from a Gaseous Source in a Porous Medium - A Field and Theoretical Comparison", pp D265-273, U.S. Geological Survey Professional Paper 650-D, (1969).

8. M.W. First, F.J. Viles Jr., P.C. Reist, D.G. Smith, D.W. Underhill and D.W. Moeller, "Annual Progress Report, Harvard Air Cleaning Laboratory," Report NYO-841-11, Oct 1967.

9. J.B. Robertson, "Behavior of Xe-133 Gas after Injection Underground: Molecular Diffusion, Materials Balance, and Barometric Pressure Effects", Report IDO-22051, July 1969.

10. Parker C. Reist, "Steady-State Measurement of Krypton-85 - Air Diffusion Coefficients in Porous Media", Environmental Science and Technology 1 (7), 566-569 (July 1976).

11. A. Boettcher, "Simple Models of Migration of Fission Gases (^{85}Kr) Through the Overlying Rocks of Underground Storage Facilities" Deutches Brennstoffinstitut, Freiberg, i.B., Sept 1972.

12. F. Reiber, "Injection of Radioactive Gaseous Effluents", Notes of Working Meeting on Ground Disposal of Radioactive Gaseous Wastes, Department of Civil Engineering, University of California, Berkeley (Jan 1965).

13. L.G. King, "Mathematical Models for Underground Injection of Gaseous Wastes into the Vadose Zone", Battelle Memorial Institute, Report BNWL-945, Dec 1968.

14. B.L. Schmultz, "Injection of Gas into the Lithosphere of the National Reactor Testing Station", Report IDO-12069, June 1969.

15. D.G. Jacobs, "Behavior of Radioactive Gases Discharged into the Ground", Nuclear Safety, 2, 175-178 (1967).

16. Paul J. Mudra, and Bruce L. Schmaltz, "An Appraisal of Gaseous Waste Disposal into the Lithosphere at the NRTS, Idaho," Report IDO-12024, Sept 1965.

17. R.M. Winar, L.E. Trevorrow and M.J. Steindler, "Feasibility of Underground Storage/Disposal of Noble Gas Fission Products", Report ANL-78-81, Argonne National Laboratory, June 1978.

18. L.E. Trevorrow et al., "Considerations Affecting Deep-Well Disposal of Tritium-Bearing Low-Level Aqueous Waste from Nuclear Fuel Reprocessing Plants," Report ANL-76-76, Argonne National Laboratory, Mar 1977.

2.8 HYDROFRACTURE

The disposal of intermediate-level waste (fission products dissolved in water) by use of hydrofracture has been performed for several years at Oak Ridge National Laboratory.[1-33] A propping agent is not usually required in the use of grout with hydrofracture. Krypton gas could be entrained to the extent of 4-6 % in the grout containing fission products just before the grout is injected into the hydrofractured medium.[1] However, no tests have been made to show that the krypton would be retained in the set, emplaced grout. Considerable engineering and care might be needed to avoid a loss of krypton at the point of mixing with possible exposures to the operators. Another variant would be to mix a krypton-loaded zeolite powder with the grout mixture just before injection. This would be operationally easier at the hydrofracture site and should give better containment of the krypton within the set grout. D. A. Knecht of the INEL has stated that stirring a fine powder of krypton-loaded zeolite with water for 10 hr produced no appreciable loss of krypton. Therefore, the water of the grout should not attack the zeolite before the grout sets within 10 hr and the water is removed as a possible reactant. A point to be proven, however, would be whether or not the alkalinity of the grout mixture would alter the zeolite and release krypton.

Either variant for hydrofracture would require study and development. The ability of set grout to withstand radiation would be limited (see section 2.3). However, the operational cost would be low because it would be shared with the cost of disposal of the other fission products. The variant with zeolite, if sucessful, could provide: (a) extremely good isolation from the public; (b) reduced effect on loss of krypton from radiation damage and appearance of rubidium due to secondary containment of solidified grout, geologic media, and depth below the earth's surface; and (c) little need for surveillance. Any variant on hydrofracture would make retrieval virtually impossible.

Other variations on the use of hydrofracture could be envisaged. For example, some fluid containing the krypton could be injected separately from the grout. The fluid with the krypton might be allowed to remain fluid or to solidify in place, e.g. by polymerization.[34-36] However, organics and water could lose much of their retentivity by radiolysis (see section 2.16.2).

REFERENCES FOR HYDROFRACTURE

1. R.M. Winar, L.E. Trevorrow, and M.J. Steindler, "Disposal of Noble Gas Fission Products Entrained in Cement Grout," in "Feasibility of Underground Storage/Disposal of Noble Gas Fission Products," Report ANL-78-81, Argonne National Laboratory, Argonne, Ill., June 1978.

2. R.E. Blanco, Oak Ridge National Laboratory, Oak Ridge, Tenn.

3. R.D. Klett, Sandia Laboratories, Albuquerque, N.M.

4. Energy Research and Development Administration, "Alternatives for Managing Wastes from Nuclear Reactors and Post-Fission Operations in the LWR Fuel Cycle", Report ERDA-76-43, Vol 4, pp 25.5 - 25.6, May 1976.

5. Oak Ridge National Laboratory, "Disposal by Hydraulic Fracturing"; pp 4-81 to 4-86, in "Siting of Fuel Reprocessing Plants and Waste Management Facilities", Report ORNL-4551, July 1970.

6. W. de Laguna et al., "Engineering Development of Hydraulic Fracturing for Permanent Disposal of Radioactive Wastes", Report ORNL-4295, Oak Ridge National Laboratory; Oak Ridge, Tenn., Aug 1968.

7. W. de Laguna, "Radioactive Waste Disposal by Hydraulic Fracturing", Nuclear Safety 11 (5), 391 (1970).

8. H.O. Weeren, "Shale Fracturing Injections at Oak Ridge National Laboratory, 1972 Series", Report ORNL-TM-4467, June 1974.

9. H.O. Weeren, "Shale Fracturing Injections at Oak Ridge National Laboratory, 1975 Series", Report ORNL-TM-5545, Aug 1976.

10. H.O. Weeren et al., "Hydrofracture Site Proof Study at Oak Ridge National Laboratory", Report ORNL-TM-4713, Nov 1974.

11. M.J. Steindler and L.E. Trevorrow, "Wastes from the Light Water Reactor Fuel Cycle", pp 55-76, in R.G. Post, ed, Waste Management 76, University of Arizona, Tucson, Ariz.

12. G.G. Eichholz; "Environmental Aspects of Nuclear Power", pp 597-599, Ann Arbor Science Publishers, Inc., Ann Arbor, Mich., 1976.

13. E.A. Merewether et al., "Shale, Mudstone and Claystone as Potential Host Rocks for the Underground Emplacement of

Waste", Report USGS-4339-5, U.S. Geological Survey, Denver, Colo., 1973.

14. W.C. McClain, "Earthquakes Induced by Underground Fluid Injection", Report ORNL-TM-3154, Oct 1970.

15. Energy Research and Development Administration, "Management of Intermediate Level Radioactive Waste, Oak Ridge National Laboratory, Final Environmental Impact Statement", Report ERDA-1533, Sept 1977.

16. W. de Laguna et al., "Safety Analysis of Waste Disposal by Hydraulic Fracturing at Oak Ridge", Report ORNL-4665, Sept 1971.

17. W. de Laguna et al., "ORNL Health Physics Division Annual Progress Report for Period Ending July 31, 1970," Report ORNL-4584, pp 5-11.

18. T. Tamura, "Disposal of Radioactive Wastes by Hydraulic Fracturing, Part IV: Chemical Development of Waste-Cement Mixes", Report ORNL-P-3002, July 1967.

19. W.C. McClain, "Hydraulic Fracturing as a Waste Disposal Method", Report ORNL-P-3054, 1967.

20. W.J. Boegly et al., "Disposal of Radioactive Wastes in Geologic Formations", CONF-660920-9, 1966.

21. H.O. Weeren, "Disposal of Radioactive Wastes by Hydraulic Fracturing, Part III: Design of ORNL's Shale-Fracturing Plant", Nuclear Engineering Design 4, 108-117 (1966), (ORNL-P-3104).

22. W. de Laguna, "Disposal of Radioactive Wastes by Hydraulic Fracturing, Part II: Mechanics of Fracture Formation and Design of Observation and Monitoring Wells", Nuclear Engineering Design 3, 432-438 (1966). (ORNL-P-1876).

23. W. de Laguna, "Disposal of Radioactive Wastes by Hydraulic Fracturing", Part I: "General Concept and First Field Experiments", Nuclear Engineering Design 3, 338-352 (1966).

24. W. de Laguna et al., "Disposal by Hydraulic Fracturing", Report ORNL-3697, pp 10-19.

25. W. de Laguna et al., "Disposal by Hydraulic Fracturing", Report ORNL-3492, pp 13-18.

26. W. de Laguna and B.L. Houser, "Disposal of Radioactive Waste by Hydraulic Fracturing", Report ORNL-2994, pp 128-136.

27. H.O. Weeren, "Evaluation of Waste Disposal by Shale Fracturing", Report ORNL-TM-5209, Feb 1976.

28. J.G. Moore et al., "Leach Behavior of Hydrofracture Grout Incorporating Radioactive Wastes", Transactions of the American Nuclear Society 23, 162-163 (1976).

29. W. de Laguna; "Radioactive Waste Disposal by Hydraulic Fracturing", Industrial Water Engineering 7 (10), 32-37 (Oct 1970).

30. R.E. Lampton et al., "Conceptual Design Report: New Hydrofracture Facility", Report ORNL-TM-4826, July 1975.

31. W.E. Tadlock, "Gas Generation by Self-Radiolysis of Tritiated Waste Materials", p 445 in Transactions of the American Nuclear Society, Winter Meeting, San Francisco, Calif., Nov 27-Dec 2, 1977.

32. K.J. Schneider and A.M. Platt, eds, "High-Level Radioactive Waste Management Alternatives, Section 1 - "Summary, Section 2 - Background and Data Base, and Section 3 - "Evaluation Methodology", Report BNWL-1900, Vol 1, Battelle Pacific Northwest Laboratories, Richland, Wash, May 1974.

33. K.J. Schneider and A.M. Platt, eds, "High-Level Radioactive Waste Management Alternatives", Section 4 - Geologic Disposal", Report BNWL-1900, Vol 2, Battelle Pacific Northwest Laboratories, Richland, Wash., May 1974.

34. R.W. Benedict, "An Evaluation of Methods for Immobilizing Solids Loaded with Krypton-85," Report ICP-1130, Idaho Operations Office, Aug 1977.

35. Y. Sousselier, "Conditioning of Wastes from Power Reactors," Transactions of the American Nuclear Society 17, 332 (1973).

36. D. Cuaz et al., "Embedding Wastes from a Light-Water Reactor in Thermosetting Resins," Transactions of the American Nuclear Society 20, 654 (1975).

37. L.E. Trevorrow et al., "Considerations Affecting Deep-Well Disposal of Tritium-Bearing Low-Level Aqueous Waste from Nuclear Fueld Reprocessing Plants", Report ANL-76-76, Argonne National Laboratory, Mar 1977.

2.9 EXTRATERRESTRIAL

J. P. Nichols and F. T. Binford wrote in 1971: "Disposal into the sun would require the development of booster systems that are considerably more reliable and inexpensive than those that presently exist or can be foreseen".[1] Since then the subject of extraterrestrial disposal of solid radioactive waste has been discussed in more detail in 1974,[2] 1976,[3] and 1978.[4,5] Malfunctions at launch still occur as witness the 17th launch of a Trident missile which ended in an explosion 5 sec after launch on December 16, 1978.[6] As with transmutation, this technique has the potential to remove the krypton-85 entirely rather than just store it, but much technology must be achieved to do it. The payload is usually considered to be put in a low-earth orbit by the Space Shuttle, then ejected to the proper destination from there using a recoverable space "tug" with or without a solar sail, for example, or ejected from an orbiting platform with an electrical accelerator. The solar sail has been recommended for impact with the sun.[7] The more readily achievable, simple use of a tug will be considered here, although the less proven variants eventually might be improvements.

A solar orbit has been considered one of the better destinations.[3,4] Any permanent container in space must dissipate its heat load by radiation. However, it might be assumed that the radiators of Burns et al[4] would keep the krypton at a tolerable temperature. Burns, et al conclude that, "The storage of nuclear waste in space does not appear to be an attractive option."[4] Nevertheless an approximate estimate of cost might be made on the extraterrestrial disposal of the 191 m^3 (STP) of krypton from the reprocessing of 2000 metric tons of nuclear fuel.[8] A favorable case will be assumed to have been attained in the form of containing the above amount of krypton in 1 m^3 of metal by ion implantation/sputtering, for example. Burns, et al[4] envision this volume of payload to be suitably shielded regarding gamma radiation and possible reentry into the earth's atmosphere. Two flights of the Space Shuttle are required and the cost of these might total about $30,000,000.[4] This is several times the estimated cost of storing 500-psi gas in cylinders in the Krypton Storage Facility. Furthermore, much more time and work would be required to demonstrate the technical feasibility of extraterrestrial disposal.

REFERENCES FOR EXTRATERRESTRIAL

1. J.P. Nichols and F.T. Binford, "Status of Noble Gas Removal and Disposal," Report ORNL-TM-3513, p 31, Oak Ridge National Laboratory, Oak Ridge, Tenn., Aug 1971.

2. K. Drumheller et al., "Extraterrestrial Disposal," Section 8, in K.J. Schneider et al., "High-Level Radioactive Waste Management Alternatives", Report BNWL-1900, Vol 4, Battelle Pacific Northwest Laboratories, Richland, Wash., May 1974.

3. R.C. Lincoln et al., "Extraterrestrial Disposal", Section 26, in J.W. Bartlett et al., "Alternatives for Managing Wastes from Reactors and Post-Fission Operations in the LWR Fuel Cycle," Report ERDA-76-43, Vol 4, National Technical Information Service, U.S. Department of Commerce, Springfield, Va., May 1976.

4. R.E. Burns et al, "Nuclear Waste Disposal in Space," NASA Technical Paper 1225, May 1978.

5. R.E. Burns and J.G. Defield, "Disposal of Radioactive Iodine in Space," NASA Technical Paper 1313, Aug 1978.

6. The Salt Lake Tribune, p 1, Dec 17, 1978.

7. J.F. McCarthy, Jr., et al., "Concepts for Space Disposal of Nuclear Waste," Massachusetts Institute of Technology, Oct 1972.

8. D.A. Knecht, "An Evaluation of Methods for Immobilizing Krypton-85", Report ICP-1125, National Technical Information Service, U.S. Department of Commerce, Springfield, Va, July 1977.

64<

BIBLIOGRAPHY FOR EXTRATERRESTRIAL

S.E. Bramer et al., "Re-entry Protection for Radioisotope Heat Sources", Nuclear Technology 11, 232 (June 1971).

R.E. Burns et al., "Nuclear Waste Disposal in Space," Report NASA Technical Paper, Dec 1977.

G.P. Dix, "Nuclear Safety of Space Nuclear Power Systems", National Topical Meeting, American Nuclear Society, p 358, Huntsville, Ala., Apr 28-30, 1970.

K. Drumheller, "Extraterrestrial Disposal of Nuclear Wastes", Nuclear Technology 24, 418 (1974).

A.L. Friedlander et al., "Aborted Space Disposal of Hazardous Material: The Long-Term Risk of Earth Encounter," Report SAI-I-RO-678-T8, Science Applications, Inc., Rolling Meadows, Ill., Feb 1977.

R.E. Hyland et al., "Study of Extraterrestrial Disposal of Radioactive Wastes, Part II: Preliminary Feasibility Screening Study of Extraterrestrial Disposal of Radioactive Wastes in Concentrations, Matrix Materials and Containers Designed for Storage on Earth", Report NASA-TM-X-68147, Lewis Research Center, Cleveland, Ohio, Oct 1972.

R.E. Hyland et al., "Study of Extraterrestrial Disposal of Radioactive Wastes, Part III: Preliminary, Feasibility Screening Study of Space Disposal of the Actinide Radioactive Wastes with 1% and 0.1% Fission Product Contamination", Report NASA-TM-X-68216, Lewis Research Center, Cleveland, Ohio, Apr 1973.

R.E. Hyland et al., "Feasibility of Space Disposal of Radioactive Nuclear Waste, I, Executive Summary," Report NASA-TM-X-2911, Lewis Research Center, Cleveland, Ohio, Dec 1973.

R.E. Hyland et al., "Feasibility of Space Disposal of Radioactive Nuclear Waste", Report NASA-TM-X-2912, Vols 1 and 2, Lewis Research Center, Cleveland, Ohio, May 1974.

W.M. Pardue et al., "Preliminary Evaluation of the Space Disposal of Nuclear Waste," Report 8-32391(100), Battelle Memorial Institute, Columbis, Ohio, Aug 30, 1977.

F.D. Postula et al., "Lightweight, Low Ballistic Coefficient, Radioisotope Fueled Capsule Prototype Development Program, Final Report," Report GA-8998, Gulf General Atomic Co., San Diego, Calif., May 1971.

F.D. Postula et al., "Lightweight, Low Ballistic Coefficient, Radioisotope Fueled Capsule Prototype Development Program, Final Report, Supplement B, Test Data and Analysis", Report GA-8998,

65<

Supplement B., Gulf General Atomic Co., San Diego, Calif., May 1971.

"Next in Space: A Commuter Special", U.S. News and World Report, Apr 22, 1974.

Anon., "Nuclear Waste Disposal Mission Analysis," Report TR-223-1874, Northrup Services, Inc., Huntsville, Ala., Nov 1977.

2.10 DISPOSAL IN THE OCEAN

Disposal of gas cylinders loaded with radioactive krypton into the ocean is another alternative. One group has concluded that ocean disposal of cylinders loaded with krypton gas would be the best method of disposal.[1,2] The London Convention on Ocean Disposal did not address specifically the question of disposal of gaseous wastes.[3,4]

The work by Laser et al.[1,2] yields a favorable conclusion concerning the technical feasibility and the low potential for exposure of humans due to ocean disposal of krypton-85. Their approach would involve lowering gas cylinders into the deep portions of the ocean from which the circulation is considered to be very low.[5,6] Pressure sensitive gas valves attached to the cylinders would admit sea water when the hydrostatic pressure would exceed a preset value. The entrance of sea water into the cylinder would presumably form crystalline krypton hydrates there;[2] also, the solubility of krypton in sea water would be increased as a result of the high pressure. It also seems logical that it would be helpful to have an added barrier such as the host metal in ion implantation/sputtering, the host zeolite, and/or some container around the contained krypton.

Evidence available in the literature shows that ocean disposal could be an acceptable disposal alternative for krypton-85. This alternative could be particularly attractive to nations which have little acceptable land for dedication to long-term radioactive waste disposal.

Table 2.10.1 lists some of the advantages and disadvantages of ocean disposal of krypton-85.

TABLE 2.10.1

ADVANTAGES AND DISADVANTAGES OF DISPOSAL

Advantages	Disadvantages
No long-term surveillance of the site is necessary.	Public acceptance of this method. Possible problems internationally with "contaminating" the ocean.
Probability of hazardous doses to humans is extremely low. (Being inert, krypton does not enter the food chain to man. Major exposure potential is from gaseous Krypton-85 in the atmosphere.)	Retrieval of Krypton would be highly impracticable.

REFERENCES FOR DISPOSAL IN THE OCEAN

1. M. Laser, H. Beaujean, J. Bohnenstingl, P. Filss, M. Heindendael, St. Mastera, E. Merz, and H. Vygen, "Off-Gas Treatment and Krypton Disposal in HTGR-Fuel Element Reprocessing", Symposium on the Management of Radioactive Wastes from Fuel Reprocessing, Nov 27 - Dec 1, 1972, Organization for Economic Cooperation and Development, Paris, March 1973; Report GERHTR-83.

2. M. Laser, "Separation, Storage and Disposal of Krypton-85," Working Paper for a technical committee of the International Atomic Energy Agency, Vienna, Austria, 1979.

3. "The Oceanographic Basis of the IAEA Revised Definition and Recommendations Concerning High-Level Radioactive Waste Unsuitable for Dumping at Sea," Report IAEA Tech-210, Report of Advisory Group Meeting, Vienna, Austria, March 21-25 1977.

4. "The Radiological Basis of the IAEA Revised Definition and Recommendations Concerning High-Level Radioactive Waste Unsuitable for Dumping at Sea," Report IAEA Tech-211, Report of Consultants Meeting, IMCO Headquarters, London, England, June 13-17 1977.

5. P. Macbeth et al., " Evaluation of Alternative Methods for the Disposal of Low-Level Radioactive Wastes," Report UC-209-03, Ford, Bacon and Davis Utah, Inc., 375 Chipeta Way, Salt Lake City, Utah 84108, Dec 1978.

6. D.R. Anderson et al., "Release Pathways for Deep Seabed Disposal of Radioactive Wastes," Report SAND74-5854, Sandia Laboratories, Albuquerque, N.M., April 1978.

69<

2.11 STORAGE ON ADSORBENTS

Charcoal is considered to be the primary adsorbent. Krypton-85 from fuel-element reprocessing could be stored by adsorption in charcoal beds.[1-3] Considerable work has been performed on off-gas cleanup using charcoal beds. Once krypton is adsorbed on the charcoal, disposal of the charcoal becomes a problem. The Division of Isotope Technology, Oak Ridge National Laboratory, has used charcoal with adsorbed krypton in 20-ft by 8-in diameter pipe at or near atmospheric pressure for about 20 yr, according to Mr. Neil Case of that division.

Table 2.11.1 indicates there are more disadvantages than advantages to using charcoal. A few potential problems with the handling and disposal of the charcoal bed with adsorbed krypton are evident:

(a) Charcoal is a ready fuel that could ignite under certain conditions. During combustion, the adsorbed krypton would be released to the atmosphere.

(b) The volume of charcoal needed to adsorb the krypton would be large compared with the volume of a zeolite (sodalite, for example) required for adsorbtion of an equal quantity of krypton.

(c) Krypton adsorbed on charcoal also appears to be more susceptible to leakage than encapsulation in zeolites.

Consequently, although charcoal may be satisfactory for short-lived fission gases, it would be inefficient and possibly hazardous for the storage of krypton-85.

TABLE 2.11.1

ADVANTAGES AND DISADVANTAGES OF KRYPTON ADSORBED ON CHARCOAL

Advantages	Disadvantages
Adsorption is a well understood technique.	Adsorption would require handling of charcoal from adsorption bed. Small particles from adsorption bed could complicate the handling problem.
Materials and process methods are available and known.	Charcoal bed could be a fire hazard.
The charcoal would have value in trapping the daughter rubidium.	Leak rates could be greater than for other alternatives such as encapsulation.
	The relatively large volume of charcoal to handle could become a problem for long-term storage.

71<

REFERENCES FOR STORAGE ON ADSORBENTS

1. Dwight W. Underhill, "Mass Transfer of Krypton-85 in Charcoal Adsorption Beds", Proceedings of the Ninth AEC Air Cleaning Conference, J.M. Morgan Jr. and Melvin W. First, eds., pp 824-847, Jan 1967.

2. D.W. Underhill, "An Experimental Analysis of Fission Gas Holdup Beds", Nuclear Science and Application 8, 255-260 (March 1970).

3. D.W. Underhill, H. Yusa and D.W. Moeller, "Preliminary Design of a Radioactive Noble Gas Adsorption Bed System for the Fast Flux Test Facility", Special Report, Harvard School of Public Health, Boston, Mass., Nov 1969.

4. R. Saito, A. Shimozato, M. Takeshima, and T. Saito, "Operating Performance of Charcoal Adsorber System Applied in BWR", Hitachi Hyoron 23, 1 (Jan 2-7, 1974).

5. Cyril M. Slansky, "Separation Processes for Noble Gas Fission Products from the Off-Gas of Fuel-Reprocessing Plants", Atomic Energy Review 9, pp 423-440.

6. G.W. Kielholtz, "Removal of Radioactive Noble Gases from Off-Gas Streams", Nuclear Safety 8 (2), 155 (Winter 1966-67).

7. W.E. Browning, "Removal of Fission Product Activity from Gases," Nuclear Safety 1 (4), 47 (1960).

8. R.D. Burnette, et al., "The Removal of Radioactive Krypton and Xenon from a Flowing Helium Stream by Fixed-Bed Adsorption", USAEC Report TID-7672, Proceedings of the Nuclear Reactor Chemistry, Second Conference, Gatlinburg, Tennessee, (Oct 10-12, 1961, 1962).

9. T.W. Leland, "Design of Charcoal Adsorbers for the HRT", USAEC Report (Oak Ridge) CF-55-9-12 (1955).

10. W.E. Browning and C.C. Bolt, "Measurement and Analysis of the Holdup of Gas Mixtures by Charcoal Adsorption Traps", USAEC Rep ORNL-2116 (1956).

11. R.E. Adams and W.E. Browning, "Fission Gas Holdup Tests on HRT Charcoal Beds", USAEC Report (Oak Ridge) CF-58-4-14 (1958).

12. W.E. Browning et al., "Removal of Fission Product Gas from Reactor Off-Gas Stream", USAEC Report (Oak Ridge) CF-59-6-47 (1959).

13. Susuma Kitani and Junichi Takada, "Adsorption of Krypton and Xenon on Various Adsorbents", Journal of Nuclear Science Technology 2 (2), 51 (1965).

14. M. Steinberg, "The Recovery of Fission Product Xenon and Krypton by Adsorption Processes", USAEC Report BNL-542 (1959).

15. J.R. Merriam et al., "Engineering Development of an Absorption Process for the Concentration and Collection of Krypton and Xenon, Summary of Progress through July 1, 1967", USAEC Report, K-1725 (1967).

16. J.R. Merriam and J.H. Pashley, "Engineering and Development for Adsorption Process for the Concentration and Collection of Krypton and Xenon, Second Summary Progress Report, July through December 1967", USAEC Report K-1745 (1968).

17. J.R. Merriam et al., "Concentration and Collection of Krypton and Xenon by Selective Absorption in Fluorocarbon Solvents", paper presented at the Tenth AEC Air Cleaning Conference in New York City, Aug 28, 1958.

18. E.J. Wilson and K.J. Taylor, "The Separation and Purification of Krypton-85 at the Multicurie Level", UK AEA Report, AERE I/R-2693 (1958).

19. M. Steinberg and B. Manowitz, "Recovery of Fission Product Noble Gases", Industrial Engineering Chemistry 51 (1), 47 (1959).

20. M. Steinberg and B. Manowitz, "An Adsorption Process for Recovery of Fission Product Noble Gases", USAEC Report BNL-493 (1958).

21. B.A. Foster, and D.T. Pence, "An Evaluation of High Pressure Steel Cylinders for Fission Product Noble Gas Storage", Report ICP-1044, Idaho National Engineering Laboratory, Idaho Falls, Idaho, Feb 1975.

22. R.A. Brown, ed., "Krypton Storage Facility: Reference Facility Description for the Recovery of Iodine, Carbon, and Krypton from Gaseous Wastes," Report ICP-1126, Idaho National Engineering Laboratory, Idaho Falls, Idaho, 1977.

23. B.A. Foster, D.T. Pence and B.A. Staples, "Long Term Storage Techniques for ^{85}Kr", Proceedings of the Thirteenth AEC Air Cleaning Conference, Report CONF-740807, pp 293-298 (1975).

24. P.L. Walker, Jr. et al., The Chemistry and Physics of Carbon, Vol 2, pp 257-371 (1966).

25. C.J. Major, B.U. Sollami and K. Kammermeyer, Industrial Engineering Chemistry, Process Design Development 4 (3), 327 (July 1965).

26. M.H. Lloyd and R.A. McNees, "Adsorption of Krypton and Xenon by Various Materials", Report ORNL-3228, Oak Ridge National Laboratory, Oak Ridge, Tenn., Nov 16, 1961.

27. R.B. Ackley and W.E. Browning Jr., "Equilibrium Adsorption of Krypton and Xenon on Activated Carbon and Linde Molecular Sieves", Report ORNL-CF-61-2-32, Oak Ridge National Laboratory, Oak Ridge, Tenn., Feb 14, 1961.

28. L.C. Lewis, "Evaluation of Adsorbents for Purification of Noble Gases in Dissolver Off-Gas", Report IN-1402, July 1970.

29. D.M. Leins, R.W. Glass, M.M. Chiles and D.J. Inman, "Monitoring and Analysis of Process Streams in a Krypton-85 Off-Gas Decontamination System", Report ORNL-TM-4923, Oak Ridge National Laboratory, Oak Ridge, Tenn., July 1975.

30. B.B. Fisher, A.E. Norris and D.G. Rose, "Adsorption of Radiokrypton on Activated Charcoal in the Presence of Hydrogen", Report LA-UR-73-1310, Los Alamos Scientific Laboratory, N. Mex.

2.12 TRANSMUTATION

R. C. Liikala et al. have outlined four minimum requirements for the successful transmutation of radioactive waste in general.[1] The process must:

(a) Consume much less electrical energy than was generated in producing the waste.

(b) Remove more noxious waste than it produces.

(c) Remove the waste much faster than natural decay would.

(d) Be practical; e.g., the krypton-85 along with significant other nuclear wastes must be removable with devices of sufficient number and capacity.

Devices not transmuting with neutrons are considered impractical.[1] The four principle types of devices are (a) charged particle accelerators producing neutrons by spallation, (b) nuclear explosives, (c) fission reactors, and (d) fusion reactors.[2] All of these may be considered as continuous devices except for (b). Nuclear explosives would employ tritium and would leave a possibly hazardous residue of this radioactive pollutant. This type of device is also unattractive politically and practically, the latter because of the many required detonations.[2]

Assessments of the practicality of transmuting krypton-85 with continuous neutrons have improved over the last few years with improvements in technology.[3-6] It is desirable to remove xenon and lower atomic-weight krypton isotopes before transmuting the krypton-85 in order to have a higher rate of reaction, to remove parasitic reactions, and particularly to prevent the formation of krypton-85 from krypton-84. The krypton-85 would need to be contained in a high concentration in something having a low parasitic absorption of neutrons, perhaps zirconium or a ceramic. Using solid krypton cooled with liquid nitrogen could improve the efficiency of the process.

It is tentatively assumed that the above requirements have been met in the following examination of the potential for transmutation in a continuous flux of neutrons. Krypton-85 contained in a fixed volume, having a concentration per unit volume of N and irradiated at a steady flux ϕ (n/cm^{2}sec) (assumed to be monoenergetic) obeys the equation:

$$dN/dt = -\lambda N - \phi N \sigma \qquad \text{(Eq 2.12.1)}$$

where,

t is the time in seconds

λ is the decay constant in sec $^{-1}$
σ is the cross section in cm^2

Since the half-life is 10.73 yr, the value of λ is 2.048×10^{-9} sec^{-1}.

J.O. Blomeke[2] gives the effective cross-section for the removal of krypton-85 as 1.5 barns in a PWR flux. The integration of Eq 2.12.1 over a campaign with a flux ϕ to a time t_w gives:

$$t_w = (\ln(N_o/N))/(\lambda + \phi\sigma) \qquad \text{(Eq 2.12.2)}$$

The above N and N_o are the final and initial values, respectively, of the krypton-85 concentration. In order to obtain the same value of N_o/N without a flux at a time $t_{w/o}$, Eq 2.12.2 is modified with $\phi = 0$ to be:

$$t_{w/o} = (\ln(N_o/N))/\lambda \qquad \text{(Eq 2.12.3)}$$

The division of Eq 2.12.2 by Eq 2.12.3 yields the relationship:

$$t_w/t_{w/o} = \lambda/(\lambda + \phi\sigma) \qquad \text{(Eq 2.12.4)}$$

Thus for any given fractional reduction in the activity of krypton-85, Eq 2.12.4 gives the relative time required with and without the flux ϕ. This quantity is plotted in Figure 2.12.1 for a PWR flux with $= 1.5 \times 10^{-24}$ cm^2. Thus a flux of 1×10^{16} n/cm^{2}sec reduces the time to 12% of the value with only decay; a flux of 5×10^{15} n/cm^{2}sec gives 21.4%; and a flux of 1×10^{15} n/cm^{2}sec reduces the time only to 57.6% of that without flux. These figures for krypton-85 are consistent with those of Blomeke for a PWR in Table 27.1 of ERDA-76-43.[2] A typical flux in a thermal fission reactor like a PWR is 3×10^{13} n/cm^{2}sec;[2] therefore, a PWR has no utility in transmuting krypton-85.

The fast flux from an LMFBR or fusion reactor, for example, would have to be greater than 10^{16} n/cm^{2}sec to be useful, because σ is low (0.087 barn) in the fast flux.* For example, with $\sigma = 0.087$ barn for an LMFBR and $N_o/N = 1000$, Eq 2.12.2 gives $t_w = 20.4$ yr for a fast flux of 1×10^{17} n/cm^{2}sec, which may be compared with Eq 2.12.3's $t_{w/o} = 107$ yr with no flux. Only a spallation device is likely to produce a continuous fast flux of 10^{17} n/cm^{2}sec according to Blomeke.[2] However, fast fluxes could be readily thermalized with beryllium or heavy water to give the same or slightly higher thermal fluxes. Hence, in Figure 2.12.1., an LMFBR could give a useful 5×10^{15} n/cm^{2}sec, a future fusion reactor in excess of 10^{16} thermal n/cm^{2}sec, and a spallation device more than 10^{17} thermal n/cm^{2}sec.

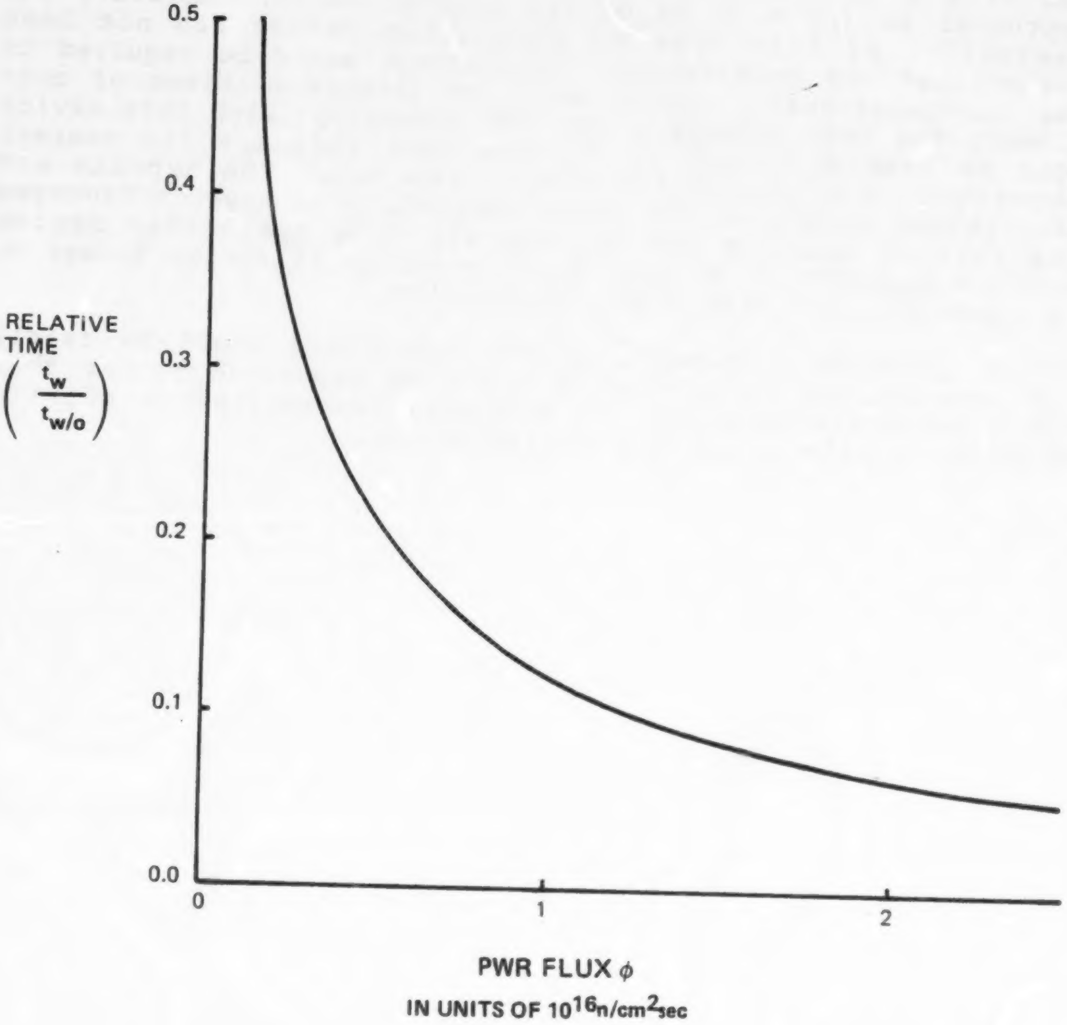


FIGURE 2.12.1 THE RELATIVE TIME IN A PWR FLUX

Consequently, the physics is most promising for a spallation device with thermalization of the neutrons. For example, a thermal flux of 1×10^{17} n/cm^{2}sec from a spallation device would require only 526 days to reduce the krypton-85 to 0.1% of its initial value (Eq 2.12.2 with $\sigma = 1.5$ barns) and a thermal flux of 5×10^{15} n/cm^{2}sec would require 3.3 yr to reduce the krypton-85 to 1/e = 36.8% of its initial value. However, the feasibility of this type of spallation device has not been demonstrated and considerable development would be required to make it operational. There would be sizable problems of containment and heat transfer, but the technology with this device might be less difficult to achieve than that for its nearest competitor, the controlled fusion reactor. The expense and radiological hazard of the prior separation of krypton isotopes plus further hazards and expense with the spallation device would be prohibitive especially considering radiation damage to the container and induced radioactivities.[7]

In principal, however, public acceptance could be fairly good because the radioactivity would be destroyed rather than stored somewhere although the probable transportation off the reprocessing site would be disadvantageous.

78<

REFERENCES FOR TRANSMUTATION

1. R.C. Liikala et al., Section 9 of K. J. Schneider and A. M. Platt, eds., "High-Level Radioactive Waste Management Alternatives," Report BNWL-1900, Vol 4, Battelle Pacific Northwest Laboratories, Richland, Wash., May 1974.

2. J. O. Blomeke et al., Section 27 of J. W. Bartlett et al., eds., "Alternatives for Managing Wastes from Reactors and Post Fission Operations in the LWR Fuel Cycle," Report ERDA-76-43, Vol 4, National Technical Information Service, U.S. Department of Commerce, Springfield, Va., May 1976.

3. J. P. Nichols and F. T. Binford, "Status of Noble Gas Removal and Disposal," Report ORNL-TM-3515, Oak Ridge National Laboratory, Oak Ridge, Tenn., Aug 1971.

4. C. W. Lindenmeier, "Transmutation Disposition," of A. M. Platt (comp.), "Advanced Waste Management Studies, Progress Report, June-July 1972," pp 21-26, Report BNWL-B-223-1.

5. W. C. Wolkenhaur et al., "Transmutation of High-Level Radioactive Waste with a Controlled Thermonuclear Reactor," Report BNWL-1772, Battelle Pacific Northwest Laboratories, Richland, Wash., Sept 1973.

6. D. Breton, "Etude Sur La Destruction Par Transmutation Neutronique Des Residus Radioactifs Provenant Du Traitement Des Combustibles Irradies," Report EUR-CEA-FC-723, Association Euratom-CEA, 92260 Fontenay-aux-Roses, France, Dec 1973.

7. R.C. Liikala et al, ibid, Appendix 9.A, Transmutation by Accelerators, Section D, Spallation Accelerators, pp. 9.A.25 - 9.A.41.

BIBLIOGRAPHY FOR TRANSMUTATION

G.A. Bartholomew, "Spallation-Type Thermal Neutron Sources," Seminar on Intense Neutron Sources, Report CONF-660925, Sept 19-23, 1966, Conference Proceedings, TID-4500, p 637.

M.J. Bell, "ORIGEN-The ORNL Isotope Generation and Depletion Code, Report ORNL-4628, Oak Ridge National Laboratory, Oak Ridge, Tenn., 1973.

J.O. Blomeke, "Partitioning and Transmutation in Perspective," presented at the Nuclear Waste Management Systems Overview Workshop, Scripps Institute of Oceanography, La Jolla, Calif., June 24-25, 1977.

H.C. Clarborne, "Neutron-Induced Transmutation of High-Level Radioactive Waste," Report ORNL-TM-3964, Oak Ridge National Laboratory, Oak Ridge, Tenn., Dec 1972.

M.V. Gregory and M. Steinberg, "A Nuclear Transformation System for Disposal of Long-Lived Fission Product Waste in an Expanding Nuclear Power Economy," Report BNL-11915, Brookhaven National Laboratory, Upton, N.Y., Nov 1967.

A.S. Kubo, "Technological Assessment of High-Level Nuclear Waste Management," ScD thesis, Department of Nuclear Engineering, Massachusetts Institute of Technology, Apr 1973.

S. Pearlstein, "Analysis of (n, 2n) Cross Section for Medium and Heavy Mass Nuclei," Nuclear Science Engineering 23 (3), 238 (1965).

S. Pearlstein, "Neutron Induced Reactions in Medium Mass Nuclei," Report BNL-16271 (1972).

M. Steinberg et al., "Neutron Burning of Long-Lived Fission Products for Waste Disposal," Report BNL-8558, Brookhaven National Laboratory, N.Y., Sept 1964.

W.C. Wolkenhaur, "The Controlled Thermonuclear Reactor as a Fission Product Burner," Report BNWL-SA-4232, Battelle Pacific Northwest Laboratories, Richland, Wash., June 1972.

W.C. Wolkenhaur, "The Controlled Thermonuclear Reactor as a Fission Product Burner," Transactions of the American Nuclear Society 15 (1), 92 (1972).

2.13 CRYOGENIC STORAGE

The radioactive krypton mixture could be stored in the condensed (solid) state by cooling with liquid nitrogen, for example.[1] The density in terms of gaseous krypton would be over 500 cm^3 (STP)/cm^3. This is one of the highest densities achievable by practical means, and would require a minimum of shielding and space oterhwise. The density of decay heat, only about 0.07 watt/cm^3 could easily be dissipated by liquid nitrogen. There is a a well-developed technology for cryogenically handling krypton.[2,4] Thus, the hazard of an ozone-hydrocarbon explosion could probably be handled. The principal hazard would be a loss of liquid-nitrogen (or other) coolant so that considerable reserve coolant capacity of some sort would be necessary. It would be worth having a large gas-tight reservoir with which to catch and recover any vaporized krypton. A final backup could be a tall stack which likely would be on the site for fuel reprocessing anyway.

One could also consider storing radioactive krypton dissolved in liquid nitrogen.

The cost of coolant for a century or so would not be high, and excellent retrievability and lack of required transportation would be in favor of this type of alternative.

REFERENCES FOR CRYOGENIC STORAGE

1. J.H. Holloway, "Noble Gas Chemistry," Methuen and Co., Ltd., London, 1968.

2. J.S. Davis and J.R. Martin, "A Cryogenic Approach to Fuel Reprocessing Gaseous Radwaste Treatment," Trans. Am. Nucl. Soc. 1978 Annual Meeting, Chicago, Ill., June 10-14, 1973, p. 176.

3. H. Yusa et al, "Application of Cryogenic Distillation to Krypton-85 Recovery," Nuclear Engineering and Design 41, 437 (1977).

4. R. v. Ammon et al, "Development of a Cryogenic Krypton-Separated System for the Off Gas of Reprocessing Plants," presented at the 15th DOE Nuclear Air Cleaning Conference, Boston, Mass., Aug 7-10, 1978, Harvard Air Cleaning Laboratory, 665 Huntington Avenue, Boston Mass. 02115.

2.14 LOW-PRESSURE STORAGE IN CAVERNS

Krypton-85 and the other noble gases from a fuel reprocessing plant could be pumped into a man-made or natural cavern.[1-5] The volume of krypton to be disposed of is 191 m^3 (STP) for 2000 metric tons of fuel,[6] and would occupy a relatively small portion of a typical natural cavern. It would be unnecessary to purify or concentrate the krypton-85 if the cavern volume were large enough to hold the off-gas at about atmospheric pressure. The gaseous mixture would have to be stored near atmospheric pressure. Otherwise it would move from the cavern along fissures in the basic rock.

Small gaseous leaks likely would occur, but they could be managed as with Porous Underground Media (see section 2.7). If moisture seeped into a cavern it would slowly dissolve the krypton and xenon (see Appendix A). However, the volume of water needed to absorb all these gases is about 15 times the volume of the gases. For caverns to be an acceptable place for gas storage, water must be maintained at low levels. This would require careful siting of the storage area and possibly special surface treatment of the cavern walls. Cementaceous coatings could help to keep the caverns dry and impede the loss of krypton, but uncertainty exists about the long-term (100-year) integrity of surface coatings.

The experience of natural gas companies in storing gas underground is helpful for assessing the possibility of storing krypton and xenon in caverns.

The advantages and disadvantages of selecting the alternative of low pressure storage in caverns are given in Table 2.14.1.

TABLE 2.14.1

ADVANTAGES AND DISADVANTAGES OF LOW-PRESSURE STORAGE IN CAVERNS

Advantages	Disadvantages
Mined-out caverns are abundant and the technique should be inexpensive.	Natural sites suitable for storage are few and difficult to find.
Acceptable sites may be located at different parts of the country.	Long-term integrity of the cavern and low transmission of gas might be difficult to assure. Small leaks would probably occur.
Migration of the gas through fissures in the unprotected cavern walls may be slow enough that it never reaches humans in high enough levels to cause a problem. Small leaks could be managed.	Krypton stored in a cavern would require much processing to recover.

REFERENCES FOR LOW-PRESSURE STORAGE IN CAVERNS

1. T. Lenahan, "There is a Place for Grouting in Underground Storage Caverns", Association of Engineering Geologists, Kansas City, Mo., Oct 24-28, 1862.

2. R.L. Nece, "Problems of Underground Storage of Wastes", Journal of Research of the U.S. Geological Survey 1, (1), 719-723 (Nov-Dec 1973).

3. M.D. Tade, "Helium Storage in Cliffside Field", Journal of Petroelum Technology, pp 885-888, July 1967.

4. Vary, Jr., et al., "The Underground Storage of Gas in the United States and Canada", 22nd and 26th Annual Report on Statistics Committee in Underground Gas Storage, American Gas Association, Dec 31, 1971 and 1976.

5. W.E. Clark and R.E. Blanco, "Encapsulation of Noble Fission Product Gases in Solid Media Prior to Transportation", Report ORNL-4473, Oak Ridge National Laboratory, Oak Ridge Tenn., 1970.

6. D.A. Knecht, "An Evaluation of Methods for Immobilizing Krypton-85," Report ICP-1125, National Technical Information Service, U.S. Department of Commerce, Springfield, Va., July 1977.

85<

2.15 LOW-PRESSURE STORAGE IN TANKS

Off-gas from the fuel reprocessing plant could be stored in low-pressure (approximately atmospheric pressure) tanks in analogy with the current practice of gas utility companies. Large, floating-roof tanks have been used for many years by utilities and oil refineries for gas storage. A similar storage alternative has been suggested previously.[1]

In this alternative, there might be no need to purify the off-gas except for corrosive impurities. Mixtures of krypton-85, krypton, xenon, etc., could be stored, although at a possibly lower concentration of krypton-85. The technology exists for large-scale storage in low-pressure tanks of non-radioactive gas. The storage of radioactive gas would require some modifications to the conventional methods. Water might not be a satisfactory barrier in this case. Modifications would include: radiation shielding, continuous surveillance of tanks for leaks, and protection from accidental impact from aircraft and other airborne projectiles. A tank of fixed gas volume having a diaphragm might be used. Unfortunately, a plastic diaphragm probably would resist radiation poorly and perhaps leak excessively.

Another version would be to bury tanks of suitable size, then remotely fill them at atmospheric pressure with the radioactive gas. Radiation shielding would be provided by the earth.

The advantages and disadvantages are listed in Table 2.15.1. This alternative would require considerably more study in order to determine if it is a realistic possibility.

TABLE 2.15.1

ADVANTAGES AND DISADVANTAGES OF LOW-PRESSURE STORAGE IN TANKS

Advantages	Disadvantages
Low-pressure storage of gases is an established practice of utilities.	Storage tanks must be protected from accidental impact such as from aircraft.
Krypton-85 would be recoverable for other uses, such as a tracer in leak detection, etc.	Fail-safe or backup containment would be required.
	Long-term site management would be required to assure tank integrity for 100-150 yr.
	Capital investment could be substantial.
	Public acceptance might be difficult to achieve.
	Does not have the same degree of permanence as certain other alternatives.
	Catastrophic failure could result in substantial doses to humans at the facility and beyond.

REFERENCES FOR LOW-PRESSURE STORAGE IN TANKS

1. H.J. Dunster and B.F. Warner, "The Disposal of Noble Gas Fission Products from the Reprocessing of Nuclear Fuel", Report AHSB (RP) R-101, United Kingdom Atomic Energy Authority, Harwell, England, Jan 1970.

88

2.16 OTHERS

2.16.1 Solutions

A possible alternative for the disposal of krypton is to dissolve the gaseous krypton in an organic material, either liquid or solid, then store this "solution." The solubility of krypton in organic materials, particularly fluorinated hydrocarbons, is much higher than it is in water.[1-3]

It has been estimated that 300 m^3 of perfluorotributylamine at atmospheric pressure could dissolve the entire krypton production from 2000 metric tons of reprocessed fuel. The solubility would be greater at higher pressure, of course. Highly perfluorinated solvents like this are denser than water[4] and, if disposed of geologically, could not be floated to the surface of the ground by rising water. Useful solubility also is obtained in terphenyl[3] which is solid at room temperature. Of the perfluoro and terphenyl types of solvents, the radiation resistance of the terphenyl type is much better known[5-6] and is very likely satisfactory. The radiolytic products from the terphenyl solvents are not unduly corrosive to containers as witness their use as a liquid coolant in reactors.[5-10] Hence the krypton-loaded, terphenyl solvent probably could be stored in an engineered structure as a solid. The radiolytic products from the perfluoro type of solvent probably would be more corrosive. Hence this type might be more promising underground as suggested by the above or perhaps packaged for disposal in the ocean (see section 2.10).

Neither of the above types of solvent is very flammable. One terphenyl type (Santowax R) melts in the range of 50°-145° C. This material, allowed to freeze in the presence of high-pressure krypton, should remain as an immobile solid having more than 1 cm^3 (STP) Kr/cm^3. Krypton could not be leached readily from this solid by water. The cost of Santowax R for 191 m^3 (STP) of krypton would be less than $200,000 at 1978 prices.

Some of the advantages and disadvantages of storing krypton in organic materials are summarized in Table 2.16.1. This extensive subject may deserve more study.

Absorbing krypton in glass has been of interest at the Battelle Pacific Northwest Laboratories[11-15] and Sandia Laboratories.[16-17] Promising results have been obtained with low-density glass, e.g. "thirsty" Vycor. The glass is both sintered and cooled under a high pressure of krypton. Useful krypton contents are believed to be obtainable and leakage rates near room temperature appear to be small.[15]

TABLE 2.16.1

ADVANTAGES AND DISADVANTAGES OF ORGANIC SOLUTIONS

Advantages	Disadvantages
Injection of the krypton-loaded terphenyl type into a well could produce an immobile solid there. Krypton - containing solvent could be stripped and the krypton made available for industrial use if liquid were stored in tanks instead of injected into the ground. Certain solvents loaded with krypton could be easily injected into porous underground media for permanent disposal.	Above-ground storage in tanks would require maintenance and surveillance which could become costly for the required storage time required. Unresearched and undemonstrated.

REFERENCES FOR SOLUTIONS

1. R.M. Winar et al., "Solubility of Krypton and Xenon in Water and Organic Liquids," Appendix C, in "Feasibility of Underground Storage/Disposal of Noble Gas Fission Products," Report ANL-78-81, Argonne National Laboratory, Argonne, Ill., June 1978.

2. W. Gerrard, "Solubility of Gases and Liquids, A Graphic Approach, Data-Causes-Prediction," Plenum Press, New York, N.Y. (1976).

3. M. Steinberg and B. Manowitz, "Recovery of Fission Product Noble Gases," Industrial Engineering Chemistry 51 (1), 47 (Jan 1949).

4. See, for example, M. Hudlicky, "Chemistry of Organic Fluorine Compounds," The Macmillan Company, New York, 1962, p. 299.

5. M. Tomlinson et al., "The Radiation and Thermal Decomposition of Terphenyls and Hydroterphenyls," presented to the IAEA Panel on the Use of Organic Liquids as Reactor Coolants and Moderators, Vienna, Austria, May 9-13, 1966, CONF-660553-2, Report AECL-2641, Atomic Energy of Canada Limited, May 1966.

6. M.M. El-Wakil, "Nuclear Energy Conversion," Ch 12, Intext Educational Publishers, San Francisco, Calif. (1971).

7. C.A. Trilling, "OMRE Operating Experience," Nucleonics 17 (11), 113 (Nov 1959).

8. E.F. Weisner, "Engineering Design of Piqua OMR," Nuclear Engineering 5 (45), 68 (Feb 1960).

9. U.S. Atomic Energy Commission, "Compilation of Organic Moderator and Coolant Technology," Report TID-7007, Parts 1 and 2, Jan 1957.

10. M. McEwen, "Organic Coolant Databook," Technical Publication AT-1, Monsanto Chemical Co., Organic Chemicals Division, July 1958.

11. G.L. Tingey, Section 3 of A.H. Platt (comp.), "Nuclear Waste Management and Transportation Quarterly Progress Report, July through Sept, 1976," Report BNWL-2179, Battelle Pacific Northwest Laboratories, Richland, Wash., Jan 1977.

12. G.L. Tingey, Section 4 of A.M. Platt (comp.), "Nuclear Waste Management and Transportation Quarterly Progress

Report," Report BNWL-2290, Battelle Pacific Northwest Laboratories, Richland, Wash., July 1977.

13. G.L. Tingey, Section 4 of A.H. Platt (comp.), "Nuclear Waste Management and Transportation Quarterly Progress Reports, April through December, 1977," Reports BNWL-2377-2, -3, and -4, Battelle Pacific Northwest Laboratories, Richland, Wash., 1977-1978.

14. W.J. Gray and G.L. Tingey, "Krypton Dissolution in Low Density Glass," of A.M. Platt and J.A. Powell (comp.), "Nuclear Waste Management Quarterly Progress Report, January through March, 1978," Report PNL-2378-1, National Technical Information Service, U.S. Department of Commerce, Springfield, Va., Aug 1978.

15. W.J. Gray et al., "Krypton Dissolution in Low Density Glass," of A.M. Platt and J.A. Powell (comp.), "Nuclear Waste Management Quarterly Progress Report, July through September, 1978," Report PNL-2378-3, National Technical Information Service, U.S. Department of Commerce, Springfield, Va. (to be published).

16. J.E. Shelby, "A Comprehensive Review of Gas Permeation, Diffusion, and Solubility in Inorganic Glasses," Report SLL-73-0259, Sandia Laboratories, Livermore, Calif., Aug 1973.

17. J.E. Shelby, Journal of Noncryst. Solids 14, 288 (1974).

BIBLIOGRAPHY FOR SOLUTIONS

W.G. Perkins et al., "Diffusion and Permeation of He, Ne, Ar, Kr and D_2 through Silicon Oxide Thin Films," Journal of Chemistry and Physics 54, 1683 (1971).

R.M. Barrer and D.E.W. Vaughn, Journal of Physical Chemistry Solids 32, 731 (1971).

L.E. Trevorrow et al., "Considerations Affecting Deep-Well Disposal of Tritium-Bearing Low-Level Aqueous Waste from Nuclear Fuel Reprocessing Plants," Report ANL-76-76, Argonne National Laboratory, Mar 1977.

93<

2.17 COMBINATIONS OF ALTERNATIVES

The emphasis in studying the preceding alternatives has been on basics rather than on combinations in order to lay a firm foundation for choosing among viable alternatives. The viable ones are then to be studied in greater depth at a later date. However, besides those alternatives identified, there also are certain combinations of those alternatives that might have synergism.

For example, the possible use of powdered zeolite loaded with krypton as an additive for the grout in hydrofracture already has been described in section 2.8. The loading limit on zeolite stated in sections 2.2 and 2.3 probably could not be increased by more than 10% beyond the original 26 cm^3 (STP)/cm^3 of bulk zeolite in the original loading process according to D. Knecht.[1] However, the bulk zeolite is about 72% void. One could add 21 cm^3 (STP) of krypton gas/cm^3 of the container's contents by also having krypton gas at 500 psi and ambient temperatures. (A sealed container also would serve a useful purpose in containing krypton leaked from the zeolite.) The total loading in the container would then be 47 cm^3 (STP)/cm^3 of contents, an improvement of 80% over that of 500 psi gas alone. A smaller container than a 23-cm-diameter cylinder probably would be required in order to maintain a maximum zeolite temperature of 150° C or less. Alternatively, the zeolite particles could be given a thin nickel coating in place to improve thermal conductivity before adding 500 psi of gas (see section 2.3). Loading the zeolite into a container for 500 psi service would probably require welding or some kind of new closure, but the improved krypton density might warrant this effort.

Similarly the loaded zeolite could, in principle, be dispersed in a matrix (section 2.3) containing krypton. However, no satisfactory matrix of this nature is known except possibly one of the terphenyl type (section 2.16.1).

REFERENCES FOR COMBINATIONS OF ALTERNATIVES

1. D.A. Knecht, "An Evaluation of Methods for Immobilizing Krypton-85," Report ICP-1125, National Technical Information Service, U.S. Department of Commerce, Springfield, Va., July 1977.

95<

3. SUMMARY OF ALTERNATIVE TECHNIQUES

The salient points of each technique described in Chapter 2 are reviewed in the following sections.

3.1 GAS AT 500 OR 2000 PSI IN CYLINDERS

The technique at 500 psi has been established during two decades of experience and has served as the basis for a study on a reference krypton storage facility (Report ICP-1126). The long-term storage probably would depend on the purity of the gas, the inherent compatibility with the liquid rubidium, and eventually on radiation damage. Corrosion-testing at the Idaho National Engineering Laboratory indicates thus far that suitable metals are obtainable for a gas cylinder and valve. The effects of radiation have not been closely examined. A pressure of 500 psi gives a density of 29 cm^3 (STP) Kr/cm^3, a value comparable with that of zeolites. Raising the pressure to 2000 psi gives a density of 100 cm^3 (STP) Kr/cm^3, but disproportionately raises the danger of sudden or gradual failure of the cylinder or valve. The design of the closure deserves particular consideration.

3.2 ZEOLITES

Current evidence indicates that 26-29 cm^3 (STP) Kr/cm^3 bulk sodalite may be obtained by loading at 15,000-30,000 psi and 400°-500°C for a few hours. The maximum leaked amount from this loaded sodalite at 150°C would be only about 0.3% of the initial amount at any time. The loaded sodalite could be stored at atmospheric pressure. The effect of krypton-85's radiation damage to sodalite has not yet been determined. The loaded sodalite is not quickly attacked by water. To avoid dusting problems, it has been proposed that the sodalite be loaded in a porous tube from which it would never be removed.

3.3 KRYPTON-LOADED SOLID SET IN A MATRIX

Nickel-coated zeolite or possibly metal matrices are the favored ones because of the lack of radiation resistance or a sufficiently low processing temperature for the others. A metal-containing matrix would facilitate a lower storage temperature of the sodalite, thus giving less leaked krypton. However, using a metal-containing matrix would entail more process complication and appreciable cost for metal, except for inexpensive ones like lead or aluminum or as yet untried, thin nickel coatings. The matrix might provide significant gamma shielding.

96<

3.4 ION IMPLANTATION/SPUTTERING

This technique has the advantage over the preceding ones of not requiring undesirable temperatures or pressures either during the loading of krypton or its storage. Furthermore, current research with amorphous alloys promises storage densities of krypton equal to or greater than those of the preceding techniques. Considerable efforts remain for development of the loading technique, compatibility with the coolant, and study of irradiation, but scale-up, leakage and probable cost do not appear to be unfavorable.

3.5 METAL VAPOR DEPOSITION

The krypton capacity at room temperature is low, only about 10 cm^3 (STP)/cm^3 of metal. The loaded metal loses krypton with the slightest attack of the metal surface. It appears to offer little advantage compared with ion implantation/sputtering.

3.6 CLATHRATES

The most promising clathrate (quinol) has disadvantages of an unfavorable combination of thermal stability and capacity which is less than 5 cm^3 (STP)/cm^3. Also, the attendant high cost of quinol is high, and the clathrate is susceptible to attack by moisture and air in the presence of radioactivity. Its advantages of ease of formation and apparent stability to radiation are outweighed by the disadvantages.

3.7 POROUS UNDERGROUND MEDIA

This technique has been successfully applied to the storage of natural gas for which the radiological and sociopolitical hazards are considerably less. The probable small leaks would be less acceptable for storage of krypton-85. A suitable porous medium would have to be near the reprocessing site, free of vertical cracks, and covered with a caprock of low permeability. Verifying the suitability of the site before use would pose difficulties. Retrieving the krypton also would be difficult. However, the technique offers the promise of low cost, the ability to manage small leaks, no need to purify the gas to be injected, and probable immunity to the effects of the rubidium daughter.

3.8 HYDROFRACTURE

The disposal of krypton-85 along with other fission products by means of hydrofracture would have the advantage of shared costs. One method would entrain krypton gas in the grout as it was being injected. Another variant would mix the krypton-loaded powder of zeolite with the grout before injection. Both methods could be limited by the low radiation resistance of the solidified grout as well as the uncertainties of hydrofracture such as possible vertical fracturing. In addition, the first method would require care not to expose workers to radioactivity at the site of hydrofracture, but remote operation would minimize this.

3.9 EXTRATERRESTRIAL

The attractiveness of the intended complete removal of krypton-85 from the earth would be tempered by the risks of launching and a conceivable re-entry into the earth's atmosphere. The cost probably would be high depending on the degree of safety that was required.

3.10 DISPOSAL IN THE OCEAN

This possibility has attracted some attention in Europe. The descent of the krypton container to the desired depth before releasing any krypton would have to be reliable. However, the solubility of gaseous krypton in sea water under a hydraulic pressure of thousands of pounds per square inch would be quite high. Alternatively, the contained krypton might be in the form of a solid (ion-implanted/sputtered metal, zeolite, in a matrix, etc.) to give more complete isolation from the biosphere.

3.11 STORAGE ON ADSORBENTS, PRIMARILY CHARCOAL

Radioactive xenon and krypton are often stored on charcoal over the short term. However, storage of krypton-85 for a century by this method would not be reliable nor efficient because of the charcoal's flammability and bulk.

3.12 TRANSMUTATION

Lower atomic-weight isotopes would have to be removed before the krypton-85 could be transmuted by neutron capture. A thermal flux would be required, but that from a PWR would be too low. The technology would be extremely difficult to achieve and the cost would be prohibitive.

3.13 CRYOGENIC STORAGE

The advantages of excellent compactness might outweigh the small continuing cost of coolant and the hazard of its possible loss. This hazard might be small with good design.

3.14 LOW-PRESSURE STORAGE IN CAVERNS

This is another inexpensive method, possibly vulnerable to small leaks, like that described in section 3.7. Due to the large volume and dilution available in some caverns, the leaks might be less serious and more easily managed.

3.15 LOW-PRESSURE STORAGE IN TANKS

The characteristics of radioactivity and krypton make this unattractive for the usual floating-roof tank. More study might be justified.

3.16.1 SOLUTIONS

Work is under way on dissolving krypton in glass. Certain organic materials such as terphenyls and perfluorinated compounds also may be useful solvents.

3.17 COMBINATIONS OF ALTERNATIVES

If loaded sodalite and 500 psi of gas were stored in the same container, the krypton density would be 80% more than that of the gas alone without necessarily incurring higher pressure.

99<

APPENDIX A

TABLES AND GLOSSARY

TABLE A.1

PROPERTIES OF NATURAL KRYPTON

Atomic weight: 83.80

Concentration in air: 1.14 ppm by volume

Melting point: -156.6° C

Boiling point: -152.3° C at 1 atm

Density at STP: 0.00374 g/cm^3

Solubility in water: 0.05 cm^3/cm^3 at 30° C and 1 atm*

Thermal conductivity: 2.09 x 10^{-5} cal/°C cm sec at 0° C

Ionization potential: 14.0 ev

---

*A. Seidell, "Solubilities of Inorganic and Metal Organic Compounds," Van Nostrand, New York, N.Y., 1940.

TABLE A.2

PROPERTIES OF NATURAL XENON

Atomic weight: 131.30

Concentration in air: 0.087 ppm by volume

Melting point: -111.9° C

Boiling point: -107.1° C

Density at STP: 0.00586 g/cm^3

Solubility in water: 0.10 cm^3/cm^3 at 30° C and 1 atm*

Thermal conductivity: 1.21×10^{-5} cal/°C cm sec at 0°C

Ionization potential: 12.1 ev

---

* A. Seidell, "Solubilities of Inorganic and Metal Organic Compounds," Van Nostrand, New York, N.Y., 1940.

GLOSSARY

Term	Definition
1 atm	1.013 x 10^5 Pa
10 atm	1.013 MPa
1 barn	10^{-24} cm^2
1 Ci	3.7 x 10^{10} d/s
ev	electron volt
INEL	Idaho National Engineering Laboratory
G	number of molecules destroyed or formed with the absorption of 100 electron-volts of radiolytic energy
1 kg	10^3g (grams)
LMFBR	Liquid Metal Fast Breeder Reactor
1 m	3.281 ft
1 mm	10^{-3} m
1 msec	1 millisecond = 10^{-3} second
n (used alone)	neutron
NRTS	National Reactor Testing Station
1 PBq	10^{15} d/s
ppm	parts per million
psi	pounds per square inch
PWR	pressurized water reactor
STP	Standard Temperature (0°C) and Pressure (1 atm) for a gas
1 TBq	10^{-3} PBq = 10^{12} d/s

THE EVALUATION OF ALTERNATIVES FOR THE ULTIMATE DISPOSAL OF KRYPTON-85

TASKS 2 AND 3

THE EVALUATION OF STORAGE TECHNIQUES AND ULTIMATE DISPOSAL

JUNE 1979

prepared for

U.S. ENVIRONMENTAL PROTECTION AGENCY
CONTRACT NO. 68-01-5001

BY

B.J. THAMER
P.J. MACBETH
D.C. RICH

FORD, BACON & DAVIS UTAH INC.
375 Chipeta Way
P.O. Box 8009
Salt Lake City, Utah 84108 . 801-583-3773

ABSTRACT

The means of immobilizing krypton-85 are compared in terms of technical feasibility, safety, relative cost and other factors such as retrievability, sociopolitical considerations, and transportational aspects. The techniques of ion implantation/sputtering and subatmospheric gaseous storage are then studied in more depth along with the reference cases of 500 psi and 2,000 psi gas in cylinders. These techniques are examined in terms of the ultimate disposition in a building, a dry well, storage in deep geological media, and ocean disposal. Specific designs are offered in several cases by way of illustration. These are described in terms of achievability, quantitative estimates of environmental hazards and comparative costs whenever possible. Finally these techniques are compared with each other in terms of the different types of ultimate disposition.

105<

CONTENTS

Chapter Title Page

ABSTRACT. ii
LIST OF FIGURES vii
LIST OF TABLES. viii

1 INTRODUCTION

1.1 BACKGROUND 1-1
1.2 OVERVIEW 1-1
1.3 ORGANIZATION OF THE REPORT 1-2

References. 1-3

2 BASIC CONSIDERATIONS FOR ULTIMATE DISPOSITION

2.1 GENERAL ASPECTS. 2-1

- 2.1.1 The Radiological Hazard 2-1
- 2.1.2 Purity of the Radiokrypton. 2-1
- 2.1.3 The Schedule of Loading, Storage and Removal 2-2
- 2.1.4 Bases for Estimates of Cost 2-2

2.2 STORAGE IN A BUILDING. 2-3
2.3 STORAGE IN A DEEP GEOLOGICAL MEDIUM. 2-3
2.4 STORAGE IN THE OCEAN 2-10
2.5 SUMMARY OF ALTERNATIVES FOR ULTIMATE DISPOSITION. 2-15

References. 2-16

3 THE APPLICATION OF THE IMMOBILIZATION TECHNIQUES TO THE DIFFERENT MODES OF DISPOSITION

3.1 STORAGE IN CYLINDERS AT 500 PSI. 3-1

- 3.1.1 Storage in a Building 3-1
  - 3.1.1.1 General Aspects. 3-1
  - 3.1.1.2 Environmental Assessment . . 3-10
  - 3.1.1.3 Estimated Cost 3-12
- 3.1.2 Storage in a Dry Well 3-13
- 3.1.3 Storage in a Deep Geological Medium. 3-15
  - 3.1.3.1 General Aspects. 3-15
  - 3.1.3.2 Environmental Assessment . . 3-15
  - 3.1.3.3 Estimated Cost 3-15

CONTENTS (Cont)

Chapter Title Page

3.1.4 Storage in the Ocean. 3-16

3.1.4.1 General Aspects. 3-16
3.1.4.2 Environmental Assessment . . 3-17
3.1.4.3 Estimated Cost 3-17

References. 3-19

3.2 STORAGE IN CYLINDERS AT 2000 PSI 3-21

3.2.1 Storage in a Building 3-21

3.2.1.1 General Aspects. 3-21
3.2.1.2 Environmental Assessment . . 3-21
3.2.1.3 Estimated Cost 3-23

3.2.2 Storage in a Dry Well 3-24
3.2.3 Storage in a Deep Geological Medium. 3-24

3.2.3.1 General Aspects. 3-24
3.2.3.2 Environmental Assessment . . 3-24
3.2.3.3 Estimated Cost 3-24

3.2.4 Storage in the Ocean. 3-25

3.2.4.1 General Aspects. 3-25
3.2.4.2 Environmental Assessment . . 3-26
3.2.4.3 Estimated Cost 3-26

References. 3-28

3.3 STORAGE AS A SOLID ALLOY 3-29

3.3.1 Storage in a Building 3-30

3.3.1.1 General Aspects. 3-30
3.3.1.2 Environmental Assessment . . 3-30
3.3.1.3 Estimated Cost 3-31

3.3.2 Storage in a Dry Well 3-33
3.3.3 Storage in a Deep Geological Medium. 3-34

3.3.3.1 General Aspects. 3-34
3.3.3.2 Environmental Assessment . . 3-34
3.3.3.3 Estimated Cost 3-35

107<

CONTENTS (Cont)

Chapter Title Page

3.3.4 Storage in the Ocean. 3-36
  3.3.4.1 General Aspects. 3-36
  3.3.4.2 Environmental Assessment . . 3-36
  3.3.4.3 Estimated Cost 3-36

References. 3-38

3.4 SUBATMOSPHERIC STORAGE 3-39

3.4.1 Storage in a Building 3-39
  3.4.1.1 General Aspects. 3-39
  3.4.1.2 Environmental Assessment . . 3-41
  3.4.1.3 Estimated Cost 3-43

3.4.2 Storage in a Dry Well 3-45
3.4.3 Storage in a Deep Geological Medium. 3-47
  3.4.3.1 General Aspects. 3-47
  3.4.3.2 Environmental Assessment . . 3-47
  3.4.3.3 Estimated Cost 3-48

3.4.4 Storage in the Ocean. 3-49
  3.4.4.1 General Aspects. 3-49
  3.4.4.2 Environmental Assessment . . 3-49
  3.4.4.3 Estimated Cost 3-49

References. 3-51

4 DISCUSSION AND SUMMARY

4.1 DISCUSSION 4-1

4.1.1 Cost. 4-1
4.1.2 Storage of Pressurized Gas. 4-1
4.1.3 Storage of a Krypton Alloy. 4-3
4.1.4 Storage of Subatmospheric Gas 4-3

4.2 SUMMARY. 4-3

108<

CONTENTS (Cont)

Appendix	Title	Page
A	COMPARISON OF IMMOBILIZATION ALTERNATIVES IN A SURFACE STRUCTURE.	A-1
	REFERENCES.	A-11
B	CALCULATIONS OF RADIOACTIVITY AND REFRIGERATION	B-1
	B.1 Radioactivity.	B-1
	B.2 Required Refrigeration	B-4
	References.	B-7
C	CALCULATIONS OF TEMPERATURE IN THE IMPLANTED ALLOY	C-1
	References.	C-2
	GLOSSARY	

109<

LIST OF FIGURES

Number	Title	Page
2-1	Mined Cavity Concept for the Disposal of Krypton-85.	2-6
2-2	Conceptual Design of Shipping Cask for Cylinders of Compressed Krypton-85.	2-8
2-3	Ocean Disposal Using a Projectile	2-14
3-1	Shielded Krypton Cask	3-3
3-2	Plan View of the KSF Near the Hot Cell.	3-4
3-3	Plan View of the KSF at First Storage Cells . . .	3-5
3-4	Elevation View of KSF near Hot Cell	3-6
3-5	Elevation View of KSF at Storage Cells.	3-7
3-6	End View of KSF Hot Cell.	3-8
3-7	End View of KSF Storage Cell.	3-9
3-8	SURF Dry Well Configuration	3-14
3-9	Subatmospheric Storage in a Building.	3-40
3-10	A Trunk Line for Subatmospheric Storage in a Building	3-42
4-1	Estimated Cost of Storage for Krypton-85.	4-2
A-1	Gas Valve	A-2
B-1	Inventory of the Krypton Storage Facility	B-3
B-2	Cumulative Emissions of Krypton-85 from KSF . . .	B-5

110<

LIST OF TABLES

Number	Title	Page
2-1	Non-Radiological Transportational Hazards during 30 Years with Cylinders of Pressurized Gas in a Geological Facility.	2-9
2-2	Summary of Non-Radiological Impacts during 30 Years of Transportation from the Reprocessing Plant to the Shipping Port.	2-12
3-1	Steady State Conditions in a KSF Cell for 500 psi Cylinders after Loss of Coolant	3-11
3-2	Steady State Conditions in a KSF Cell for 2000 psi Cylinders after Loss of Coolant.	3-22
3-3	Steady State Conditions in a Cell with Implanted Radiokrypton after Loss of Coolant. . .	3-32
3-4	Steady State Cell Conditions for Subatmospheric Storage after Loss of Coolant	3-44
A-1	Summary of Immobilization Techniques for ^{85}Kr in a Surface Structure.	A-3
B-1	Final Radiological Impact on World's Population for Thirty Years of Loading 15,000,000 Ci ^{85}Kr per Year into the KSF.	B-6

1. INTRODUCTION

1.1 BACKGROUND

Krypton-85 is the only noble-gas fission product of importance that remains a few months after fission. It has a half life of 10.73 years and exists principally in the elemental state like the other noble gases. Unless it is immobilized, the radiokrypton moves almost entirely into the earth's atmosphere where it becomes uniformly distributed within a very few years. The radiological hazard to the world's population is due mostly to the whole-body dose from the human immersion in the earth's atmosphere.[1] Heretofore the reprocessing of nuclear fuel has resulted in venting krypton-85 to the atmosphere through a stack. However, the U.S. Environmental Protection Agency has promulgated a radiation-protection standard to the effect that the krypton-85 entering the atmosphere from reprocessing should not exceed 50,000 Ci per 1,000 megawatt-year of electrical energy.[2] The rule is to be effective January 1, 1983, even though commercial reprocessing does not exist at present.

Spent fuel representing the annual output of 67 nuclear reactors of 1,000 megawatts each (roughly the current U.S capacity) has been estimated by D. A. Knecht to contain 17,000,000 Ci of krypton-85.[3] Since it is estimated that a facility for reprocessing radiokrypton would only recover about 90 percent of it because of some "down time", at issue is the immobilization and storage of about 15,000,000 Ci per year.[4,5] This will be taken in this report to represent the nominal, annual amount of storable radiokrypton in this country. The new EPA emission limit would permit no more than 22 percent of this amount to escape and half of this would presumably be emitted during the down time of the reprocessing facility for radiokrypton.

1.2 OVERVIEW

The purpose of this study is to examine the means to immobilize and store krypton-85 until it has decayed to comparatively innocuous levels. Task 1 of the study was a survey of the literature and led to a report listing alternatives.[1] The balance of the work is contained in this report. The seventeen techniques of immobilization provided in Reference 1 have been rated in Appendix A according to technical feasibility, safety, comparative cost and other factors. Limitations of time have prevented the further study of all promising techniques of immobilization. Hence only two, ion implantation/sputtering and subatmospheric storage in tanks, have been chosen for further evaluation, along with the reference cases of 500 and 2,000 psi gas in cylinders. These four techniques are examined here in relation to the principal alternatives for ultimate disposition, namely storage in a building, a dry well, a geological repository or in the ocean. The various combinations are evaluated in terms of achievability, radiological and

112<

non-radiological hazards, and comparative cost. The chosen values of gas pressure during storage are only intended to be illustrative.

1.3 ORGANIZATION OF THE REPORT

Chapter 2 begins by discussing the radiological hazards of krypton-85, the purity of the product to be stored, storage schedules, and bases of cost. It then describes the generic characteristics of geological and ocean disposal. Chapter 3 discusses all combinations of the principal immobilization techniques and disposal locations. Hazards and comparative cost are estimated. Chapter 4 briefly discusses the relative values of the different combinations and states major conclusions of the study. Appendix A describes how the immobilization techniques to be studied in detail were selected from the original seventeen. Appendix B gives calculations of radioactivity and refrigeration requirements. Calculations of heat transfer in a krypton alloy make up Appendix C.

REFERENCES FOR CHAPTER 1

1. B.J. Thamer et al, "The Evaluation of Alternatives for the Ultimate Disposal of Krypton-85, Task 1 - A Compilation of Alternatives," Report UC-261-01, Ford, Bacon & Davis Utah, Inc., Feb 1979.

2. U.S. Environmental Protection Agency, "Environmental Radiation Protection Standards for Nuclear Power Operations," Federal Register 42, No. 9, Title 40, Part 190 (1977).

3. D.A. Knecht, "An Evaluation of Methods for Immobilizing Krypton-85," Report ICP-1125, National Technical Information Service, U.S. Dept. of Commerce, Springfield, VA, July 1977.

4. "Environmental Impact Statement: Management of Commercially Generated Radioactive Waste," Report DOE/EIS-0046-D, Department of Energy, Washington, DC, April 1979.

5. "Technology for Commercial Radioactive Waste Management," Report DOE/ET-0028, Department of Energy, Washington, DC, 1979.

2. BASIC CONSIDERATIONS FOR ULTIMATE DISPOSITION

2.1 GENERAL ASPECTS

Storage of krypton-85 might take place in a building, a cavern within the earth, or at the bottom of the ocean. Use of a dry well for storing krypton-85 is also touched on in this report. Most other locations are less favorable. For example, extraterrestrial disposal has been judged to be impractical (Appendix A).

A number of considerations must be addressed in order to consistently compare the efficacies of the ultimate modes of disposition mentioned above. These are discussed in the sections that follow.

2.1.1 The Radiological Hazard

Krypton-85 produces its most serious effects in humans by means of whole-body exposure.[1,2] The local exposure of individuals to released radiokrypton can be estimated by well-known methods.[3,4] However, if radiokrypton is released through a tall stack, the principal health effects are usually world wide, as the radiokrypton becomes uniformly distributed throughout the world's atmosphere within one or two years. This is expected to be the more common mode of exposure. It would add to, and perhaps eventually exceed, the still sizable exposure due to radiokrypton from the testing of nuclear weapons.[5] If 15,000,000 Ci of krypton-85 were exhausted through a tall stack, the world-wide commitment would be 8,800 man-rems of whole-body exposure or 7 health effects.[6] Reprocessing and storage of radiokrypton would still probably result in some loss of krypton-85 to the atmosphere from the reprocessing plant and the storage facility.

2.1.2 Purity of the Radiokrypton

The containment of krypton-85 as a gas at pressure may enhance corrosion due to the presence of the rubidium daughter of krypton-85, as well as non-inert impurities of the gas. Oxygen-containing impurities would probably be the most serious in a krypton-85 product from cryogenic distillation because of their radiolysis to corrosive ozone.[7] Fluorine- and chlorine-containing impurities could have serious effects in a product from selective absorption in a fluorocarbon solvent.[8] The Idaho National Engineering Laboratory is conducting tests on general corrosion, pitting, embrittlement, and, to some extent, the effects of impurities.[9] Tests up to 400°C for 5,000 hours in liquid rubidium showed no effect on stressed monel 400, alloy A286, type 316 or type 347 stainless steel. However, a more oxided sample of rubidium has given significant attack at higher temperatures.

Teflon threading tape should probably be avoided in the storage cylinder or tank and the associated valves and tubing connections. The reason is that radiolysis of polytetrafluoroethylene would lead to an attack on the metal in the threads and this, in turn, might harm the seal. It is preferrable to weld all connections, provided sound, ductile welds can be obtained.

The interaction of intense radiation with non-inert-gas components in stored gas would make the gas corrosive toward a metallic container. For example, the very reactive gas ozone would be produced by radiolysis of oxygen impurities or any component containing oxygen.[7,10] This radiation enhancement of corrosion would be most serious from the beta radiation because 99.57 percent of krypton-85's disintegration is by beta decay.[11] It would be prudent to reduce the non-inert-gas components of the radioactive krypton to a concentration below 0.01 percent. It will be assumed here that this purity has been attained before the storage of radioactive krypton in the gaseous form. It will also be assumed that the inside of any metallic storage container for the gas be essentially free of impurities such as rust. The latter could be a source of oxygen and also interfere with the seating of a valve. The added cost to achieve these requirements would be comparatively small for storing cylinders of 500 psi gas in the Krypton Storage Facility (KSF), a conceptual, surface-storage facility assumed to be located at the site of a fuel reprocessing plant.[12] The greater durability of the metallic container and its closure could repay the added cost.

2.1.3 The Schedule of Loading, Storage and Removal

The nominal, annual production of radiokrypton from power reactors amounts to about 169 m^3 (STP) of total krypton gas, of which 94 percent is nonradioactive. The Krypton Storage Facility and other disposal locations analyzed here are considered to be loaded at a fixed rate of 15,000,000 Ci/yr for a loading period of 30 years, as indicated in Figure B-1 of Appendix B.[13,14] The containers are to be stored for a storage time of 20 years beyond the 30-yr loading period. If the containers are retrieved, they are serially removed, oldest first, at the same rate that they were originally loaded. This will take an additional thirty years. At that time the radioactivity in each container will have decayed to 4 percent of its initial value. The container's contents might then be dispersed through a tall stack.

2.1.4 Bases for Estimates of Cost

Comparative costs are estimated for each combination of immobilization technique and storage location, to the extent possible. Cost estimates are in 1979 currency unless otherwise stated. An annual inflation rate of 7 percent is assumed whenever appropriate. Architect-engineer fees are included.

The estimated accuracy range for KSF capital costs was given as 30 percent in Ref. 14. The accuracy range of this report's costs for storage is estimated to be 40 percent, based on the latter figure and other uncertainties. Any large error in the cost estimate for a particular facility is likely to be repeated to some degree in the estimate for the others. Therefore, comparisons of cost are more meaningful than the accuracy range would suggest.

2.2 STORAGE IN A BUILDING

Storing tanks of gaseous radiokrypton in a building would have the advantages of ready surveillance and remedial action as well as secondary containment in the form of negative-pressure concrete cells. The only designed facility of this nature appears to be one specifically for 42.5-l cylinders containing 500 psi gas and sometimes called the Krypton Storage Facility (KSF).[12-14] This design is described in detail in Section 3.1.1.

2.3 STORAGE IN A DEEP GEOLOGICAL MEDIUM

The deep geological medium that has received the most attention for storing solid radioactive waste in this country is bedded salt as at the Waste Isolation Pilot Plant in southeast New Mexico.[15] It is sometimes said that no one type of medium is currently considered to be superior to its competitors for solid waste.[16] This generalization is for solid waste, the container of which may be corroded away without necessarily exposing humans to radiation. Exception should be taken for krypton-85, in at least its gaseous form. A corrosion-caused leak of a cylinder or tank of radioactive krypton gas could lead rather quickly to exposures at the surface from krypton-85. Hence the corrosiveness usually attributed to geological salt is a disadvantage for present purposes compared with alternate media. The situation is less clear in the case of ion implanted/sputtered krypton in an alloy. Research is needed to show how easily the alloy with its cap of nickel, for example, would be corroded to yield gaseous krypton. For gaseous krypton, however, favorable deep geological locations may eventually be found among media such as the basalt near Richland, Washington,[17,18] tuff, granite or argillite at the Nevada Test Site,[19] or others of interest to the Office of Nuclear Waste Isolation.[20] It may be a matter of a few years before definitive results are obtained with these media. However, for the present, this group will be considered to be as favorable as geological salt for the storage of krypton gas.

Geological storage has often been viewed as sealing off waste in underground chambers with no intention of identifying or remedying leaking containers. This would be a poor means of ultimate disposal for pressurized gas cylinders because experience predicts the occurrence of some leakage and need for remedial action. The leaked radiokrypton would diffuse to the surface of the earth. More experience is needed to find whether or not stored radiokrypton from ion implantation/sputtering or subatmospheric tanks would be sufficiently leakproof for a century or so to be safely sealed in the above manner. A capability for remedial action in the geological facility might remove the above restrictions. A facility of this nature has been suggested for the Nevada Weapons Testing Site using existing tunnel systems.[21] It would involve venting a concrete tunnel containing the radioactive waste in order to provide convective cooling. The hazard of contaminating the atmosphere at the ground level would be considerable even with double containers around pressurized radiokrypton. Furthermore the weak cooling would probably limit the density of storage within the concrete tunnel except when subatmospheric tanks are used.

Geological cavities are subdivided into natural caverns and mined ones. Mined cavities are more readily adapted to the present purpose and have been selected as the reference example for this comparative analysis. Mines considered include both existing nonproductive mines and new excavations made explicitly for krypton disposal. The mined-cavity concept incorporates excavated rooms and tunnels or chambers in a geologic formation for disposal of wastes. Disposal in geologic cavities has been previously proposed and presented in great detail.[22-25] As applied specifically to the disposal of krypton, the mined cavity repository in a geologic formation would be of the room-and-pillar type. A typical Mined Cavity Disposal Facility (MCDF) would consist of a number of excavated rooms and connecting tunnels located below the surface of the ground, in a geologic formation selected on the basis of favorable characteristics to contain radioactive waste. The total excavation would cover up to 4 acres of floor space on one or several levels, as required by the emplacement methods considered advantageous to safe disposal operations. The excavations would be appropriately vented to a stack while the cavity was occupied. Conventional drilling and mining operations to excavate new disposal rooms would be controlled to avoid compromise of previously completed sections of the MCDF.

Sedimentary basins in the continental interior and coastal plain that contain salt might be suitable for mined chambers at depths of about 910 m or less.[22] Salt might be suitable because of its low permeability, high thermal conductivity, and natural plasticity. The corrosion of the containers might be lessened by mounting them on racks above the floor of the salt chamber. The Salina Formation, the interior province of the Gulf Coast dome region, and the Paradox Basin are all considered to be formations of high potential. Salt formations have received the most attention in the past with respect to the disposal of high-level radioactive wastes.

Clay and shale formations also may be suitable as disposal sites because of their low permeabilities. The greatest potential for using shale is in arid and semi-arid parts of the

United States, where chambers can be mined well above existing water tables. The basin and range province of the western United States (particularly the great basin exclusive of seismic risk zone 3) appears to have potential for mined cavities above deep water tables in tuff, shale, or argillite.

The stable continental interior where sedimentary cover is thin or absent, the shield area of the North Central states, and the metamorphic belt of the Eastern United States (primarily the Piedmont) are also possibly suitable for mined cavities. Rock formations with low permeability at depths from 305 m to 6,100 m within these areas may be suitable for mined chambers.[22] Some granitic deposits (for example, the Climax Stock at the Nevada Test Site) may also be suitable.[19] Rocks originating from consolidation of materials ejected by volcanic eruptions (tuffs) are sometimes dense and compact enough to be appropriate. The lower thermal conductivity of shale, clay, and granite is a drawback in comparison to salt. However, the non-salt media are comparatively noncorrosive. The thermal conductivity of granite is half that of salt at room temperature, but it decreases less at high temperatures.

A parameter of importance in geological disposal without forced cooling is the thermal loading, i.e., the number of kilowatts of heat generated by the waste, per acre of repository. A safe figure for salt may conservatively be 50 kW/acre of total area of cavity, above which there is concern for the mechanical properties of the salt.[24] Certain other media such as granite have lower thermal conductivities than salt, but have mechanical properties that are less sensitive to heat. This question of allowable thermal loading is still under study. However, a limiting thermal load of 50 kW/acre will also be assumed for granite in subsequent sections. It is believed to be a conservative figure.

Shafts or tunnels would connect the excavated rooms to surface receiving facilities for introducing packaged waste and operations personnel into the MCDF central control area. From this point, the waste would be transferred through tunnels to its point of disposal in excavated rooms.

Surface receiving and handling facilities would occupy a few hundred square meters and would be the only visible surface evidence of the MCDF except for a stack. The krypton would be delivered to the facility by rail. Temporary storage space and capacity for container repair would be provided within the facility. The receiving facility could be operated to permit use of the surrounding surface land even though the land would be above the underground repository.

A conceptual sketch of the mine concept is shown in Figure 2-1.[25] Entrance to the mine may be by vertical shafts, when the excavated rooms are below the entrance, and by horizontal tunnels when the mine is excavated into a hill. Details of

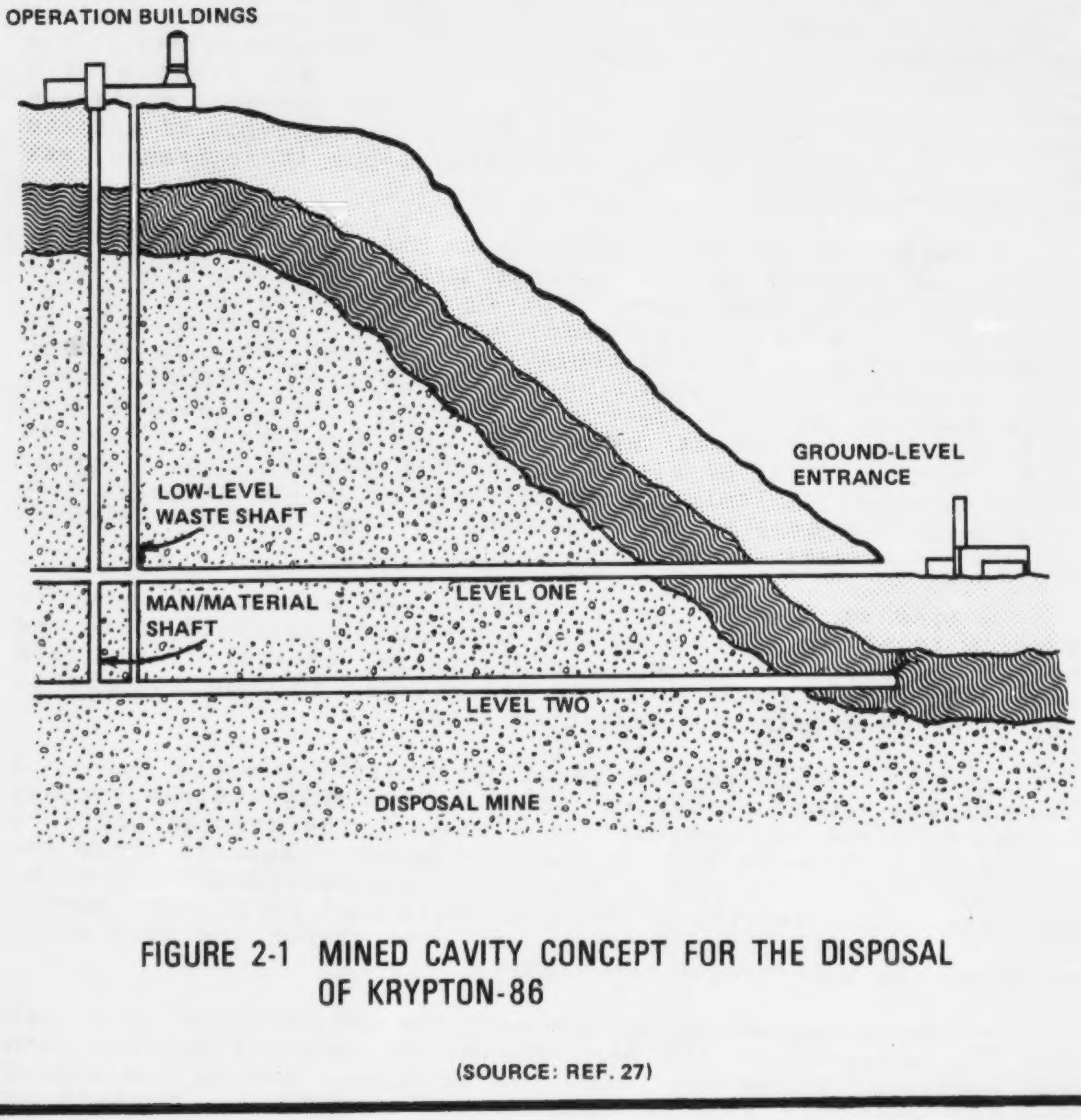

OPERATION BUILDINGS
GROUND-LEVEL ENTRANCE
LOW-LEVEL WASTE SHAFT
MAN/MATERIAL SHAFT
LEVEL ONE
LEVEL TWO
DISPOSAL MINE

FIGURE 2-1 MINED CAVITY CONCEPT FOR THE DISPOSAL OF KRYPTON-86

(SOURCE: REF. 27)

120<

waste disposal operations in a reference MCDF would depend upon the immobilization technique. Some of them are discussed in Chapter 3. Care would be taken to select sites where competing uses of nearby natural resources would not compromise the long-term suitability for waste disposal. Vertical shaft sites are more common than suitable horizontal shaft locations.

The cavern alone may not give the protection to the public that a concrete cell of the KSF would due to the upward leakage and diffusion of krypton.[26,27] The MCDF could provide as much protection to the public from krypton-85 as an above-ground building such as the Krypton Storage Facility if the containers of radiokrypton were contained in leak-proof concrete cells and the geological caverns themselves were vented to a tall stack.

Non-krypton waste disposal in mined cavities has taken place in West Germany.[28] The mining technology is well developed. In fact, it is better developed than the techniques of immobilizing krypton-85. The geological disposal of radioactive waste has been seriously retarded by public opposition in West Germany. Opposition like this for solid waste together with the obvious volatility of krypton make sociopolitical acceptance in this country seem questionable unless very good containment has been demonstrated.

The cost of transportation to the site of the geological storage is to be considered in comparing costs with an above-ground building such as the Krypton Storage Facility. For example, a KSF would very likely be on the same site as the reprocessing plant that would produce the krypton-85, but a typical site of geological storage could well be hundreds of miles away. These factors are taken into account in Chapter 3. Building something like a storage cell, for example, in a geological cavern is somewhat more expensive than building the same unit at the earth's surface. This factor also is included in Chapter 3.

A water-cooled shipping cask has been proposed by J.O. Blomeke and J.J. Perona[29] (Figure 2-2). There would be six casks to a rail car. A maximum credible accident could be described as the sudden loss of the entire contents of the contained cylinders, although the shipping cask is designed to withstand accident conditions. This would result in approximately 312,000 Ci being released into the air from three 500 psi cylinders. Assuming the pressurized radiokrypton to be diluted 10,000 fold, the concentration of radioactivity would be 216 Ci/m^3. A person remaining in this cloud for 10 seconds would receive a whole body dose of 0.09 rem and a skin dose of 20 rem. These are within current limits for the annual dose to an individual in a restricted area.[30] However, a similar release of radioactivity within the confines of the geological storage could cause doses which exceed the annual limit.

Of the non-radiological hazards, those of transportation to

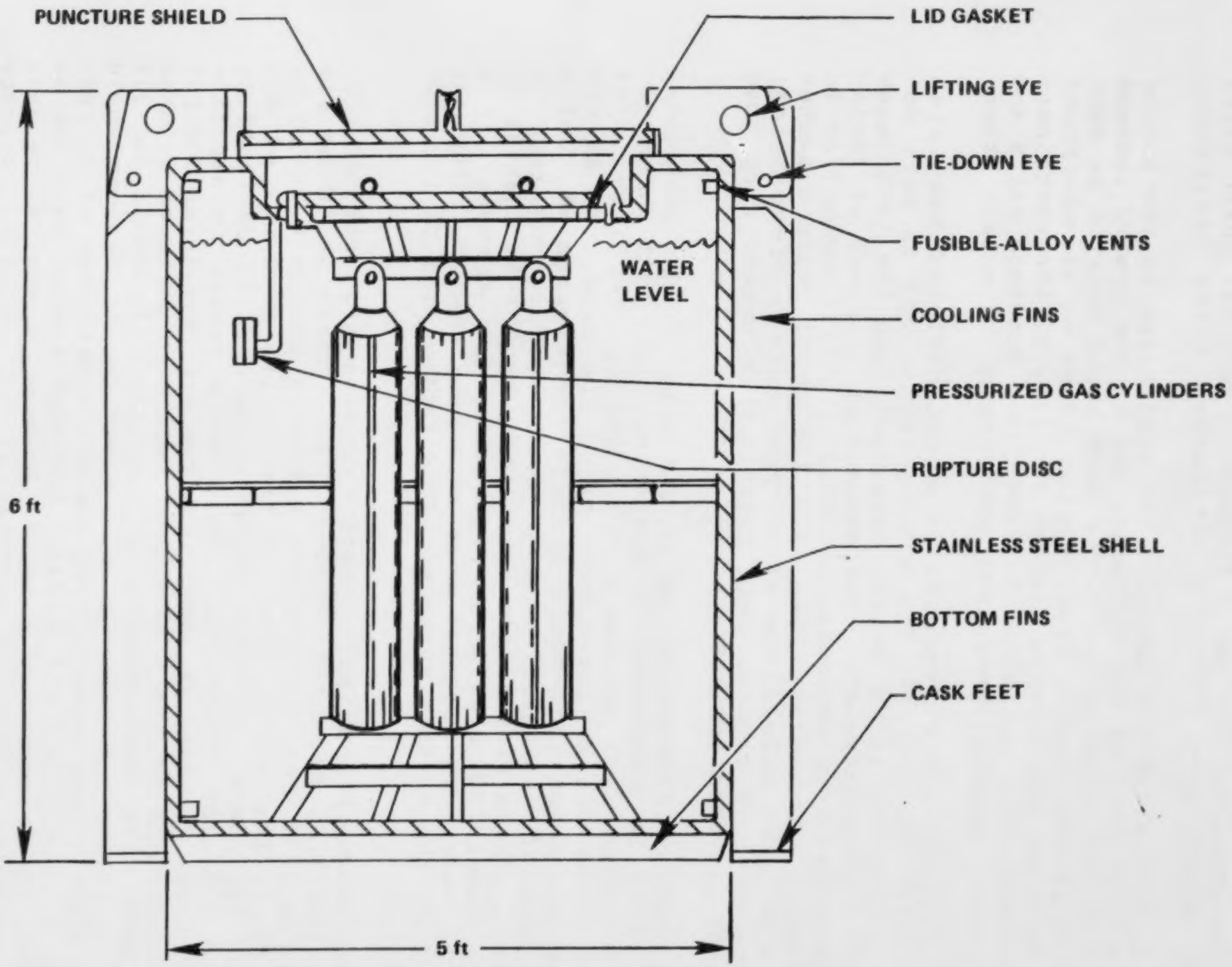

PUNCTURE SHIELD
LID GASKET
LIFTING EYE
TIE-DOWN EYE
FUSIBLE-ALLOY VENTS
WATER LEVEL
COOLING FINS
PRESSURIZED GAS CYLINDERS
RUPTURE DISC
STAINLESS STEEL SHELL
BOTTOM FINS
CASK FEET
6 ft
5 ft

FIGURE 2-2 CONCEPTUAL DESIGN OF SHIPPING CASK FOR CYLINDERS OF COMPRESSED KRYPTON-85

(SOURCE: REF. 31)

TABLE 2-1

NON-RADIOLOGICAL TRANSPORTATIONAL HAZARDS DURING 30 YEARS WITH CYLINDERS OF PRESSURIZED GAS IN A GEOLOGICAL FACILITY[31-34]

	Eastern Site	Western Site
Average Transit Distance (mi) to Disposal Site	1,000	2,000
Total Train Car Distance (mi)	230,000	460,000
Total Projected Accidents	0.03	0.07
Total Projected Injuries	0.09	0.2
Total Projected Fatalities	0.02	0.03

and from the facility are considerably more than those of the consruction or operational phase of the facility itself. Table 2-1 summarizes the estimates of hazards for two repositories serving regions of the United States[33-36] for 500 psi cylinders, three to a cask, or 2000 psi cylinders, one to a cask.

In summary, geological salt has received the most attention up to now but several other media are being currently studied. The latter may have an advantage in being less potentially corrosive, although the containers of the radiokrypton will not contact the geological medium if stored on racks. Geological storage as an alternative to storage in a building would have the advantage of comparative immunity to tornadoes, hurricanes and flying objects, but could be subject to the deleterious effects of inward leaking moisture.

2.4 STORAGE IN THE OCEAN

The possibility of disposing of containerized radiokrypton in the ocean rests partly upon the growing body of knowledge with respect to the ocean disposal of solidified waste. Oceans offer the advantage of large distances from human populations and are attractive to nations that are unable or unwilling to consecrate parts of their land to the disposal of radioactive waste.

The disposal of radioactive waste in the ocean is not currently practiced by the United States. However studies continue, principally for mid-plate/mid-gyre locations at water depths greater than 4000 m and latitudes less than about 50°.[35] One option, the emplacement in the seabed, has been studied for solidified waste.[36,37] It might also be applicable to radiokrypton contained either in a solid by ion implantation/sputtering or as high-pressure gas in a strong container. The International Atomic Energy Agency would provisionally limit the annual ocean disposal of krypton-85 to 10,000,000 Ci at a single site and ten times that for an entire water body with 10^{17} m^3 of seawater (a small ocean).[38] However, the IAEA recommends more detailed calculations for this matter.

Satisfactory resistance to corrosion by seawater might be obtained with nickel plating for implanted alloys or a nickel container for the gas. Nickel would have the advantage that it would resist alkali hydroxide which would be produced when seawater reaches the rubidium daughter of the krypton-85. A high-nickel alloy might be used in place of nickel for improved strength of the pressurized container if the resistance to corrosion would not suffer too much. The cost of the nickel in these containers would not be important compared with the cost of most options. Each container could be surrounded by 3 to 4 in of cast lead, lead eutectic or other gamma-absorbing seawater-resisting metal to protect the container from radiolytically enhanced corrosion by seawater.

A pressurized cylinder emplaced in the seabed would be maintained upright by the projectile that would contain it. The water pressure would be greater than the gas pressure. If the eventual inward leakage of seawater were arranged to be at the lower end of the cylinder, the gas would still remain captive and might become stabilized as solid krypton hydrate at the 1-2°C and more than 5,000 psi pressure of the seawater.[39] This high pressure could benefit heat transfer, to prevent nucleate boiling of the seawater at depths greater than 2000 m.

At the present time there are three good sites where a possible reprocessing plant could be located. These are at Barnwell, S.C., on the east coast and Hanford, Washington, and Idaho National Engineering Laboratory (INEL) in the west. These three sites would require shipping ports, possibly Savannah River and Seattle. Since the majority of the present and proposed nuclear power plants are located in the eastern half of the United States, in the calculations that follow it is assumed that 75% of the krypton generated at reprocessing plants would be obtained at Barnwell. It is further assumed that 12.5% would be obtained at each of INEL and Hanford. Thirty years of transportation (Section 2.1.3) from the reprocessing plants to the shipping ports would result in the projected number of accidents listed in Table 2-2, assuming that 6 casks can be transported on each train car. As can be seen, the probability of even one accident during the 30-year period is very small. Routine transportation doses would occur only during transit to the seaport. They would be 0.1 man-rem/169 m^3 (STP) radiokrypton based on transport by train.

The transport ship would require a shielded room where the cylinders could be removed from the shiping casks by remote control and either placed in a projectile or dropped directly into the ocean. Its arrival at the disposal site should be confirmed by two independent locating techniques--perhaps one employing sonar detection and the other using satellite measurements.

It seems preferrable to exclude seawater from the radiokrypton as long as possible rather than implement its entrance with a valve. The use of a valve on a pressurized container might be justified only if the radiokrypton were to be retrieved from the bottom of the ocean. However, the expense and complication of some such arrangement seems unjustified and it will be assumed that retrieval is not sought. A gas cylinder would have to be strong to withstand a seawater pressure of more than 6,000 psi at depths of 4,000 m or more. Hence a cylinder of DOT specification 3AA6000(6K size) holding 42.5 l is suggested as being more suitable than one of specification 3AA2400 because of the higher (internal) pressure rating. A gas pressure of 2,000 psi rather than 500 psi would be beneficial in helping the cylinder to withstand the hydraulic pressure. Leakage from whatever source would probably first occur at the end of the cylinder containing the valve or closure. Seawater would enter

TABLE 2-2

SUMMARY OF NON-RADIOLOGICAL IMPACTS DURING 30 YEARS OF TRANSPORTATION FROM THE REPROCESSING PLANT TO THE SHIPPING PORT

(Source: Refs. 31 through 34)

	Barnwell, S.C.	Hanford, Wash.	INEL, Idaho	Total
Transit Distance (mi)	50	150	750	
Train-Car Miles (total)	22,500	13,500	67,500	
Projected Accidents	0.0036	0.0018	0.0096	0.015
Projected Injuries	0.0096	0.0051	0.0255	0.040
Projected Fatalities	0.00072	0.00039	0.00195	0.0031

126<

the container but the gas would remain inside if the "closure" were pointing downward. Hence this orientation of a pressurized cylinder should be arranged in ocean disposal. It would be automatically obtained if the cylinder were so oriented in a free-dropping projectile of the type envisioned for solid radioactive waste.[25]

Figure 2-3 represents a projectile with five cylinders of the 6K size. (A smaller projectile to accommodate only one cylinder might be better operationally, but it would require further design and development.) Each cylinder would be properly supported. As the projectile falls, a valve would allow the ready ingress of seawater to the outermost surfaces of the cylinders. Thus the implosion of the projectile into the cylinders would be prevented and one would have excellent passive cooling of the cylinders from then onward. The dimensions of the projectile could be modified to give optimum performance[25] and, if desired, the pressurized cylinders could be replaced with the hollow ones from ion implantation/sputtering. Refitting the ship would cost about $3,650,000 for navigation equipment[25] and $950,000 for shielding and remote control. Each projectile would cost about $1300. The volume of cargo would be composed principally of the loaded shipping casks and projectiles. The costs of land transportation would average about $700 per loaded cask and $185 per projectile. Both the nautical and land costs of transportation could be taken to scale approximately with the volume of cargo.[25]

The radiokrypton disposed of in the above type of projectile would be difficult to retrieve for unauthorized purposes. The depth of the ocean and the penetration into the sediment would make it difficult to reach the material.

The sediments into which the radiokrypton penetrates and the projectile itself provide additional barriers to the movement of krypton-85. Solid krypton hydrate would probably form before the captive gaseous krypton of a gas cylinder would loose krypton as a gas. The gaseous krypton would be entirely consumed in forming the krypton hydrate. However, it is estimated that the solubility of the krypton hydrate in the seawater is about 1.7 l (STP)/l seawater.[39,40] Hence the krypton would not be lost as a gas but as material dissolved in seawater diffusing into the seawater of the sediment. The movement of seawater within sediment beds has been measured to be very low and abyssal seawater of a mid-plate/mid-gyre region circulates very slowly to the surface of the ocean. The degree of transport by organisms would have to be proven.

In summary, the principal challenges of ocean disposal are to contain the radiokrypton in the primary container at the bottom of the ocean as long as possible, then exploit the secondary containment from a high solubility in the sediment and slow movement of the abyssal water from the mid-plate/mid-gyre region. The primary containment would likely be breached at the

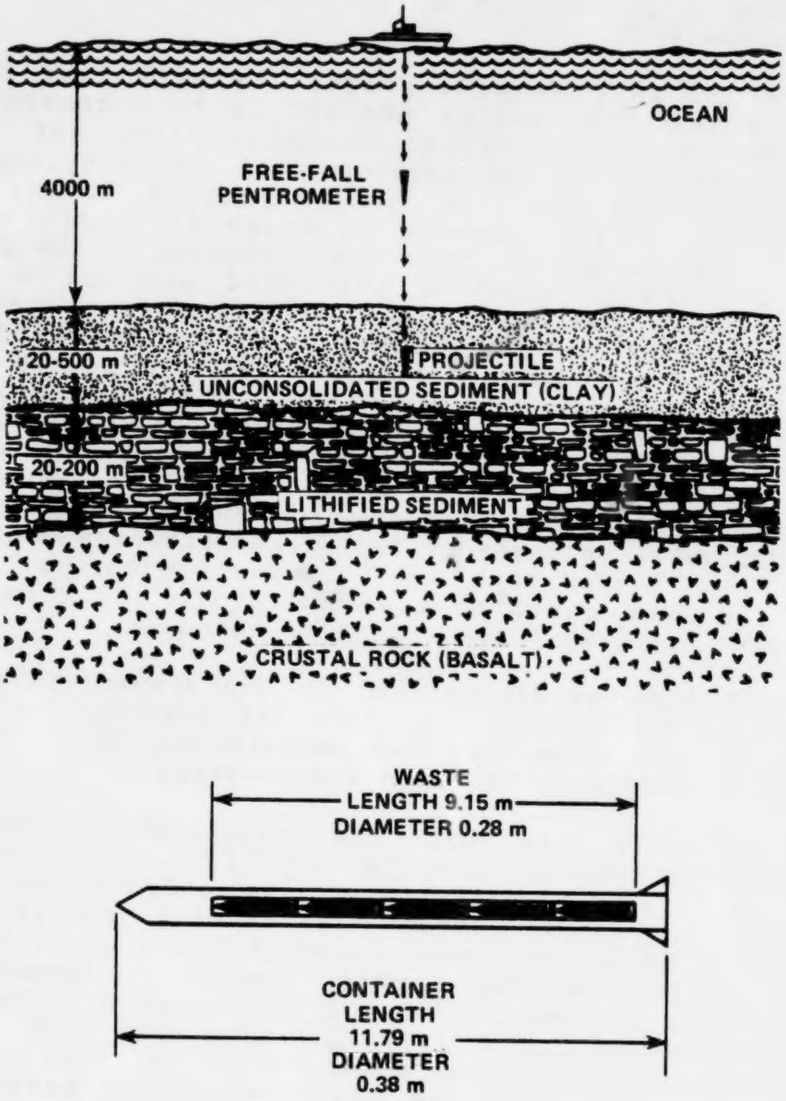

OCEAN
4000 m
FREE-FALL
PENTROMETER
20-500 m
PROJECTILE
UNCONSOLIDATED SEDIMENT (CLAY)
20-200 m
LITHIFIED SEDIMENT
CRUSTAL ROCK (BASALT)
WASTE
LENGTH 9.15 m
DIAMETER 0.28 m
CONTAINER
LENGTH
11.79 m
DIAMETER
0.38 m

FIGURE 2-3 THE USE OF A PROJECTILE

(SOURCE: REF. 27)

128<

valve or closure of a pressurized cylinder due to corrosion. It it believed that a welded closure would be more resistant than a valve in this situation.

2.5 SUMMARY OF ALTERNATIVES FOR ULTIMATE DISPOSITION

The principal locations for the ultimate storage of the radiokrypton are considered to be in a building, a geological cavern, or a mid-plate/mid-gyre region of the ocean. Storage in a building would be the most expensive and vulnerable to tornadoes, flying objects and sabotage. However, it would have the capability for excellent surveillance and remedial action as well as probably requiring no transportation from the site of the reprocessing plant that produces the radiokrypton. A geological cavern usually would provide insufficient secondary containment for a gaseous radioactivity so that its use in this capacity should be restricted to immobilization techniques that have very low emissions from the primary container. Pressurized gas cylinders would not fall in this category, but subatmospheric tanks might. Both geological storage and ocean disposal would usually require transportation off the site of the reprocessing plant. In contrast to the other locations, the krypton would not ordinarily be retrievable from ocean disposal. Ocean disposal would also probably be the least expensive of the three modes of ultimate disposition.

Chapter 3 will briefly consider using dry wells for disposal. This mode may require no off-site transportation, could allow surveillance and remedial action, and may be less vulnerable to tornadoes, flying objects, and sabotage than a building.

REFERENCES FOR CHAPTER 2

1. U.S. Environmental Protection Agency, "Environmental Analysis of the Uranium Fuel Cycle, Part III-Nuclear Fuel Cycle" Report EPA-520/9-73-003-D, U.S. Environmental Protection Agency, Criteria and Standards Division, Washington, D.C.

2. U.S. Environmental Protection Agency, "Environmental Analysis of the Uranium Fuel Cycle, Part IV--Supplementary Analysis--1976," Report EPA 520/4-76-017, U.S. Environmental Protection Agency, Office Radiation Programs, Washington, D.C.

3. U.S. Environmental Protection Agency, "Environmental Radiation Protection Standards for Nuclear Power Operations," Federal Register 42, No. 9, Title 40, Part 190 (1977).

4. D.H. Slade, ed., "Meteorology and Atomic Energy 1968," Report TID-24190, National Technical Information Service, U.S. Dept. of Commerce, Springfield, VA, Mar 1976.

5. W.S. Diethorn and W.L. Stockho, "The Dose to Man from Atmospheric ^{85}Kr," Health Physics 23, 653 (1972).

6. B.J. Thamer, et al, "The Evaluation of Alternatives for the Ultimate Disposal of Krypton-85, Task 1 - A Compilation of Alternatives," Report UC-261-01, Ford, Bacon & Davis Utah Inc., Feb 1979.

7. M.C. Sauer, Jr., "Pulse Studies of Gases," pp. 112-116 of M. Burton and J.L. Magee, "Advances in Radiation Chemistry," Vol. 5, John Wiley and Sons, New York, 1976.

8. W.D. Burch et al, "Retention of Gaseous Fission Products in Reprocessing LMFBR Fuels," Report IAEA-AG-63-2, from advisory group meeting on reprocessing of Liquid Metal Fast Breeder Reactor Fuels, Leningrad, USSR, May 17-21, 1976, International Atomic Energy Agency, Vienna, Austria, May 1976.

9. T.R. Pinchback and D.A. Knecht, "The Development of Process and Storage Materials Suitable for Krypton-85 Waste Management," paper presented at the International Symposium on Science Underlying Radioactive Waste Management, Boston, Mass., Nov 29-Dec 1, 1978.

10. J.W.T. Spinks and R.J. Woods, "An Introduction to Radiation Chemistry," John Wiley & Sons, New York, 1964.

11. A.B. Christensen, "Physical Properties and Heat Transfer Characteristics of Materials for Krypton-85 Storage," Report ICP-1128, Idaho Operations Office, Idaho Falls, Idaho (1977).

12. R.A. Brown et al, eds., "Reference Facility Description for the Recovery of Iodine, Carbon and Krypton from Gaseous Wastes," Report ICP-1126, Idaho Operations Office, Idaho Falls, Idaho, Apr 1978.

13. Department of Energy, "Environmental Impact Statement: Management of Commercially Generated Waste," Report DOE/EIS-0046-D, Dept. of Energy, Washington, D.C., 1979.

14. Department of Energy, "Technology for Commercial Radioactive Waste Management," Report DOE/ET-0028, Dept. of Energy, Washington, D.C., 1979.

15. W.D. Weart, "Geotechnical Evaluation of the WIPP Site in Southeast New Mexico," p. 217 in R.G. Post, ed., proceedings of the symposium, "The State of Waste Disposal Technology and the Social and Political Implications," Feb 26 - Mar 1, 1979, Waste Management 79, Tucson, Arizona.

16. F. Donath, "Technical Issues in Waste Isolation," p. 145 in R.G. Post, ed., proceedings of the symposium, "The State of Waste Disposal Technology and the Social and Political Implications," Feb 26 - Mar 1, 1979, Waste Management 79, Tucson, Arizona.

17. Staff of Basalt Waste Isolation Program, "Basalt Waste Isolation Program, Annual Report - Fiscal Year 1978," Report RHO-BWI-78-100, Rockwell Hanford Operations, Energy Systems Group, Richland, WA, Oct 1978.

18. Chemical and Engineering News, April 16, 1979, p. 7.

19. Nevada Operations Office, "Nevada Nuclear Waste Storage Investigations," U.S. Dept. of Energy, Las Vegas, Nev, Jan 1979.

20. U.S. Dept. of Energy, "ONWI Library Reports List," Report ONWI-2, 2nd. ed., Division of Waste Isolation, U.S. Dept. of Energy, Washington, D.C., Nov 15, 1978.

21. R.P. Hammond, "Nuclear Wastes and Public Acceptance," American Scientist 67, 146 (1979).

22. U.S. Energy Research and Development Administration, "Alternatives for Managing Wastes from Reactors and Post-Fission Operations in the LWR Fuel Cycle," Report ERDA-76-43, May 1976.

23. A. Platt, et al, "High-Level Radioactive Waste Management Alternatives," Report BNWL-1900, May 1974.

24. C.M. Koplik, R.J. Byrne, M.S. Giuffre, J.W. Bartlett, et al, "Information Base for Waste Repository Design," Report NUREG/CR-0495, Vols. 1-7, The Analytic Sciences Corporation, Reading, MA 01867, Mar 1979.

25. P. Macbeth et al, "Evaluation of Alternative Methods for the Disposal of Low-Level Radioactive Wastes," Report NUREG/CR-0680, Ford, Bacon & Davis Utah, Salt Lake City, Utah, Apr 1979.

26. J.B. Robertson, "Diffusion from a Gaseous Source in a Porous Medium--A Field and Theoretical Comparison," U.S. Geological Survey Professional Paper 650-D, pp. D265-73, 1969.

27. J.B. Robertson, "Behavior of Xenon-133 Gas after Injection Underground: Molecular Diffusion, Materials Balance, and Barometric Pressure Effects," Report IDO-22051, July 1969.

28. "Proceedings of the International Symposium on the Management of Radioactive Wastes from the Nuclear Fuel Cycle," Report IAEA-SM-207, International Atomic Energy Agency, Vienna, 1976.

29. J.O. Blomeke and J.J. Perona, "Management of Noble-Gas Fission Product Wastes from Reprocessing Spent Fuels," Report ORNL-TM-2677, Oak Ridge National Laboratory, Oak Ridge, TN, November 21, 1969.

30. Code of Federal Regulations, Title 10, Standards for Protection against Radiation, Part 20, January 1, 1976.

31. "Environmental Survey of Transportation of Radioactive Materials to and from Nuclear Power Plants," Report WASH-1238, December 1972.

32. "Final Environmental Statement on the Transportation of Radioactive Material by Air and Other Modes," Report NUREG-0170, December 1977.

33. H.K. Elder, et al, "Assessment of the Risk of Transporting Spent Nuclear Fuel by Truck," Report PNL-2588, Battelle Pacific Northwest Laboratory, Richland, WA.

34. "Accident Facts - 1977 Edition," National Safety Council, IL, 1977.

35. International Atomic Energy Agency, "The Radiological Basis of the IAEA Revised Definition and Recommendations Concerning High-Level Radioactive Waste Unsuitable for Dumping at Sea," Report of Meeting Held June 13-17, 1977, Reports IAEA-210 and -211, International Atomic Energy Agency, Vienna, 1978.

36. D. R. Anderson, et al, "Release Pathways for Deep Seabed Disposal of Radioactive Waste," Report SAND74-5854, April 1978.

37. D. R. Anderson, ed., "The Third International Seabed High-Level Waste Disposal Assessment Workshop, Albuquerque, N.M., February 6-7, 1978; A Report to the NEA Radioactive Waste Management Committee," Report SAND78-0369, Oct 1978.

38. International Atomic Energy Agency, "The Radiological Basis of the IAEA Revised Definition and Recommendations Concerning High-Level Radioactive Waste Unsuitable for Dumping at Sea," Report IAEA-211, p 46, International Atomic Energy Agency, Vienna, August 1978.

39. J. H. Holloway, "Noble Gas Chemistry," Methuen & Co., Ltd., London (1968).

40. D. Wood and R. Caputi, "Solubilities of Krypton and Xenon in Fresh and Salt Water," Report USNRDL-TR-988, U.S. Naval Radiological Defense Laboratory, San Francisco, CA, April 1966.

3. THE APPLICATION OF THE IMMOBILIZATION TECHNIQUES TO THE DIFFERENT MODES OF DISPOSITION

3.1 STORAGE IN CYLINDERS AT 500 PSI

For over 20 years the Idaho National Engineering Laboratory and its predecessor have extracted krypton-85 from the reprocessing of nuclear fuel and bottled it in nominally 50-liter cylinders for shipment to Oak Ridge National Laboratory in shipping casks. The cylinders have been limited to an initial pressure of 500 psi and contents of 74,000 Ci of krypton-85.[1] The valve has been modified to be entirely metallic with a valve seat of monel and a valve stem of Type 303 stainless steel. General experience has also been generated for over 50 years in the storage of various nonradioactive hazardous gases in similar cylinders.[2] As discussed in Section 2.1.2, the intense radiation of krypton-85 activates gas impurities, notably those containing oxygen, to increase the hazard of corrosion. Hence it is assumed that impurities other than xenon be reduced to the level of 0.01 percent for storage at 500 psi.

3.1.1 Storage in a Building

3.1.1.1 General Aspects

The storage of pressurized cylinders of krypton-85 in a building has been the subject of engineering design.[3-6] The radioactive krypton obtained at the reprocessing plant is considered to be bottled in 42.5-l cylinders at 500 psi.[5,6] A cylinder having 104,000 Ci of krypton-85 generates 151 watts, which leads to a wall temperature of about 50°C.[2] The Idaho National Engineering Laboratory is studying the most suitable alloys for the cylinder and its all-metal valve.[7] It might well reduce the possibility of leakage if some threaded connections at the valve were replaced by welds. However, obtaining ductile welds might require expensive materials or annealing procedures.

A high radiation field in the gas would enhance the corrosiveness of oxygen-containing impurities toward a storage cylinder or tank. The enhanced corrosion would be more serious for 500 psi storage than subatmospheric storage because both the gas concentration and radiation intensity would be at least 30-fold greater at 500 psi. Hence it is considered prudent to have oxygen-containing impurities below 0.01% for 500 psi but less restrictive (0.1%) for subatmospheric storage.

The storage building is assumed to be nearby on the same site as the reprocessing plant that generates the krypton-85. A shielded cask similar to that of Figure 3-1 would be used to transfer filled cylinders to the Krypton Storage Facility (KSF).[3] The lead shielding adjacent to the cylinder would be 3 in. thick. The weight of the loaded shipping cask would be

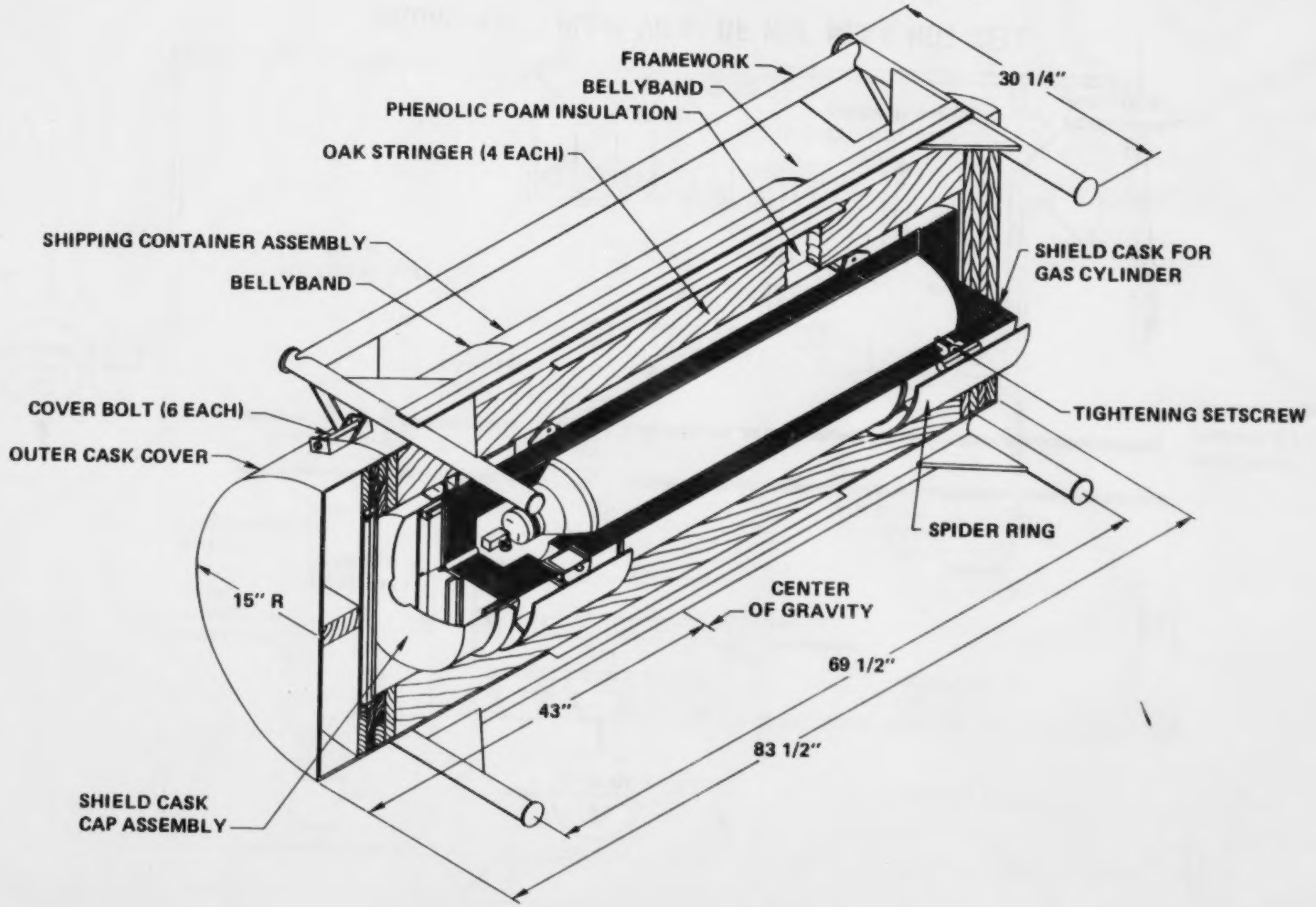
5200 lb. The shielded cask would enter the KSF (Figures 3-2 through 3-7) near the hot cell, where the cylinder would be checked for leakage and a protective cap would be installed. (Alternatively the radiokrypton might be piped to the hot cell for bottling there.) Cylinders already in storage would be periodically removed and transferred to the hot cell (Figures 3-2 and 3-6) for examination. The contents of a leaker would be transferred to a clean, empty cylinder.

The KSF is assumed to receive 169 m^3 (STP) of radiokrypton or 145 cylinders per year.[5,6] A loading period of three decades followed by storage and eventual removal of containers has been described in Section 2.1.3. Three similar storage areas are thus envisaged, each containing 14 cells.[5,6] Operating personnel would be protected against krypton-85 in accordance with government standards.[8] The transport of a cylinder between the hot cell and a storage cell would be via a type of fork lift on a central track (Figures 3-2 and 3-3). Each cell would be sized to take 104 cylinders (Figures 3-3, 3-5 and 3-7). Each cell would be shielded with lead and would have isolation valves. Decay heat would be removed by closed-cycle, down-draft air. Leakage within a cell would be detected by monitors.

Personnel would enter the KSF storage area to install or remove cylinders in the storage cell. The monitoring and exposures to radiation would generally be closely controlled. An overhead bridge crane of 125 ton capacity (Figure 3-6) would be available for the service and maintenance of each storage cell in the KSF. It might be noted that the DuPont study envisions remote operation for handling the cylinders of krypton-85.[4] The DuPont study considers the krypton-85 product to be derived from selective absorption in difluorodichloromethane. If so, the impurities and corrosive behavior might well be different from the krypton-85 product derived from the cryogenic distillation assumed in the design of the KSF.

Both the DuPont and KSF designs envisage the air of the krypton storage cells to be at a slight negative pressure. Each cell of the KSF would have krypton-85 detectors, with alarms, in its ventilation ducts. The hot cell of the KSF would also have a radiation monitor and alarm. There would be a full complement of radiation-detecting instruments and alarms for personnel areas. There would be a diesel generating unit for emergency power.

The air of a KSF cell would be completely recovered in case a cylinder was found to be leaking badly. The isolation valves in the ventilation system of the storage cell would be opened to bypass the chiller and flush the cell's atmosphere to remove the ^{85}Kr-contaminated air. The removed air could be returned to the reprocessing plant for the extraction of krypton-85. A less desirable alternative would be to bottle the air under pressure for storage of contaminated air. The large amounts of bottled



FRAMEWORK
BELLYBAND
PHENOLIC FOAM INSULATION
OAK STRINGER (4 EACH)
SHIPPING CONTAINER ASSEMBLY
BELLYBAND
COVER BOLT (6 EACH)
OUTER CASK COVER
15" R
SHIELD CASK CAP ASSEMBLY
30 1/4"
SHIELD CASK FOR GAS CYLINDER
TIGHTENING SETSCREW
SPIDER RING
CENTER OF GRAVITY
69 1/2"
43"
83 1/2"

FIGURE 3-1 SHIELDED KRYPTON CASK
(SOURCE: REF. 3)

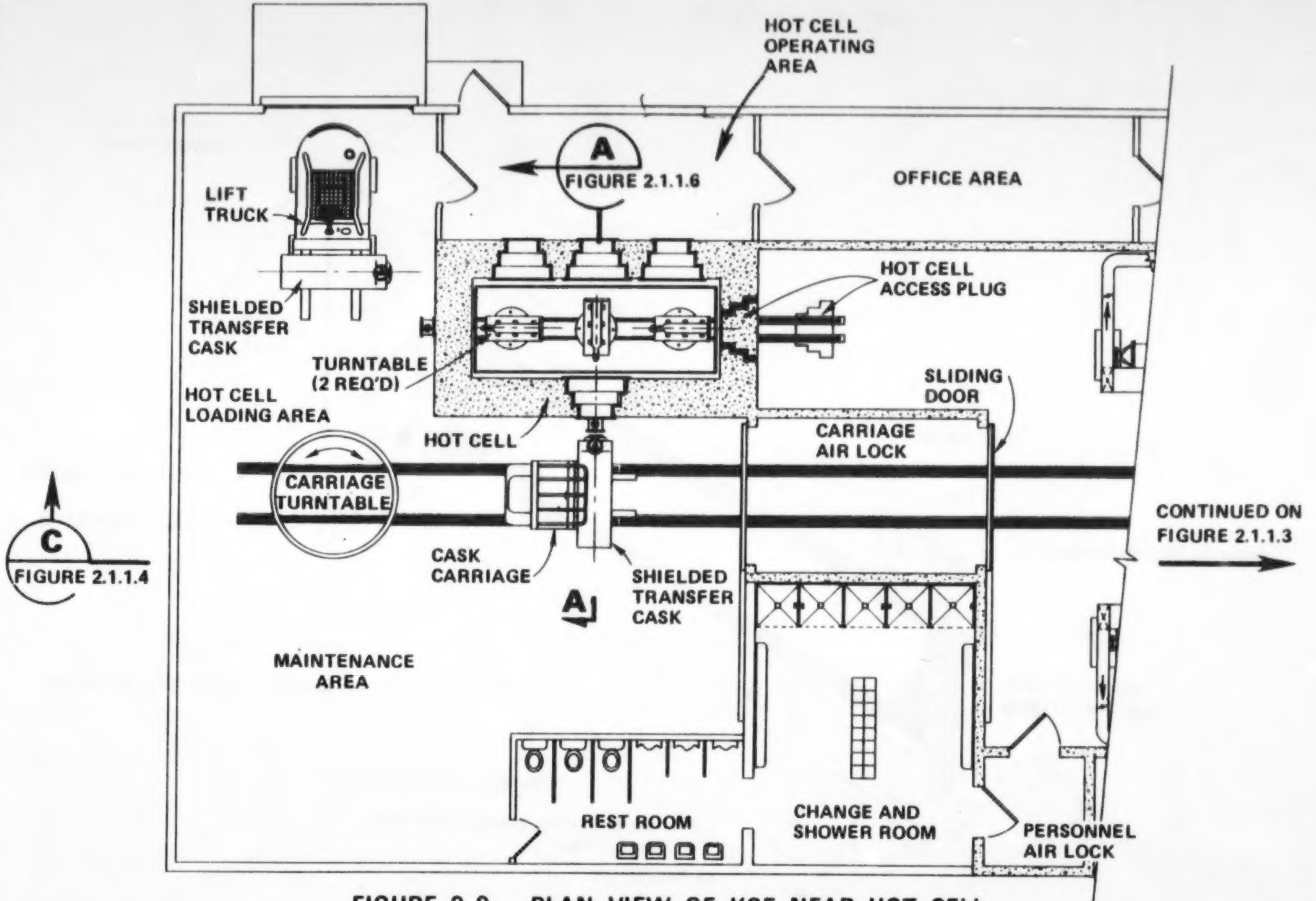

HOT CELL OPERATING AREA
A
FIGURE 2.1.1.6
OFFICE AREA
LIFT TRUCK
SHIELDED TRANSFER CASK
HOT CELL ACCESS PLUG
TURNTABLE (2 REQ'D)
SLIDING DOOR
HOT CELL LOADING AREA
HOT CELL
CARRIAGE AIR LOCK
CARRIAGE TURNTABLE
C
FIGURE 2.1.1.4
CONTINUED ON FIGURE 2.1.1.3
CASK CARRIAGE
SHIELDED TRANSFER CASK
A
MAINTENANCE AREA
REST ROOM
CHANGE AND SHOWER ROOM
PERSONNEL AIR LOCK

FIGURE 3-2 PLAN VIEW OF KSF NEAR HOT CELL

(SOURCE: REF. 3)

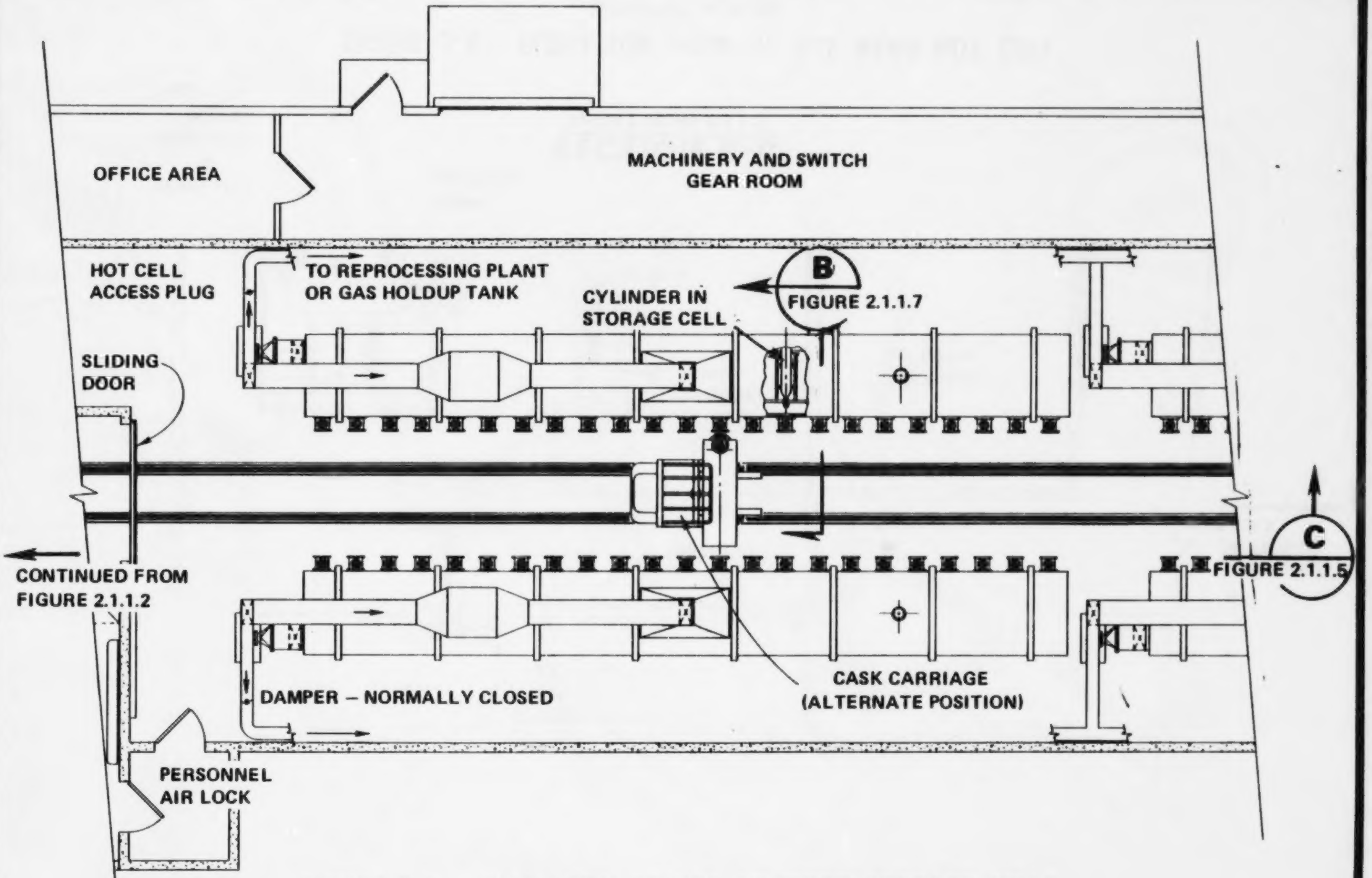

OFFICE AREA
MACHINERY AND SWITCH GEAR ROOM
HOT CELL ACCESS PLUG
TO REPROCESSING PLANT OR GAS HOLDUP TANK
CYLINDER IN STORAGE CELL
B
FIGURE 2.1.1.7
SLIDING DOOR
CONTINUED FROM FIGURE 2.1.1.2
C
FIGURE 2.1.1.5
DAMPER – NORMALLY CLOSED
CASK CARRIAGE (ALTERNATE POSITION)
PERSONNEL AIR LOCK

FIGURE 3-3 PLAN VIEW OF KSF AT FIRST STORAGE CELLS

(SOURCE: REF. 3)

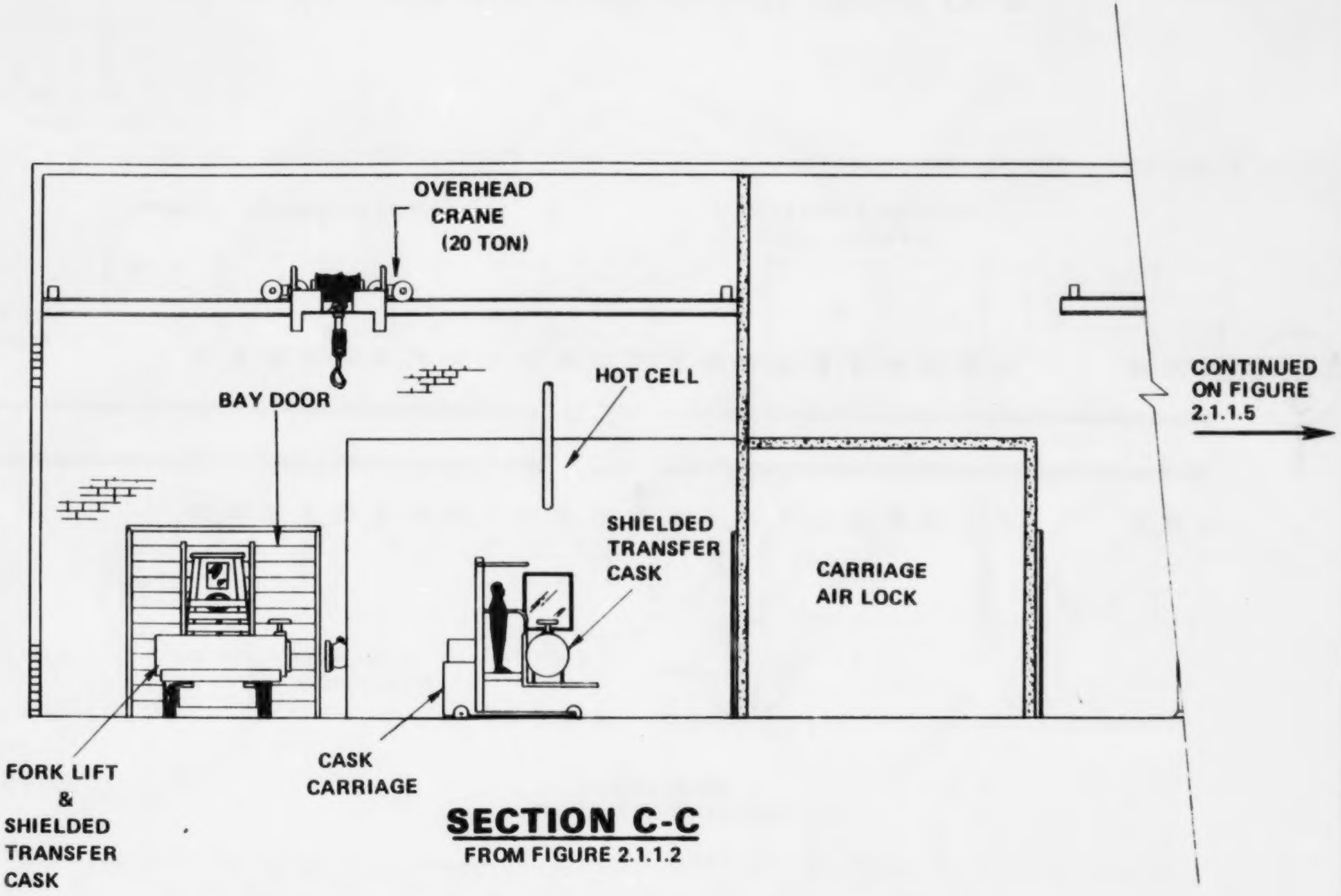
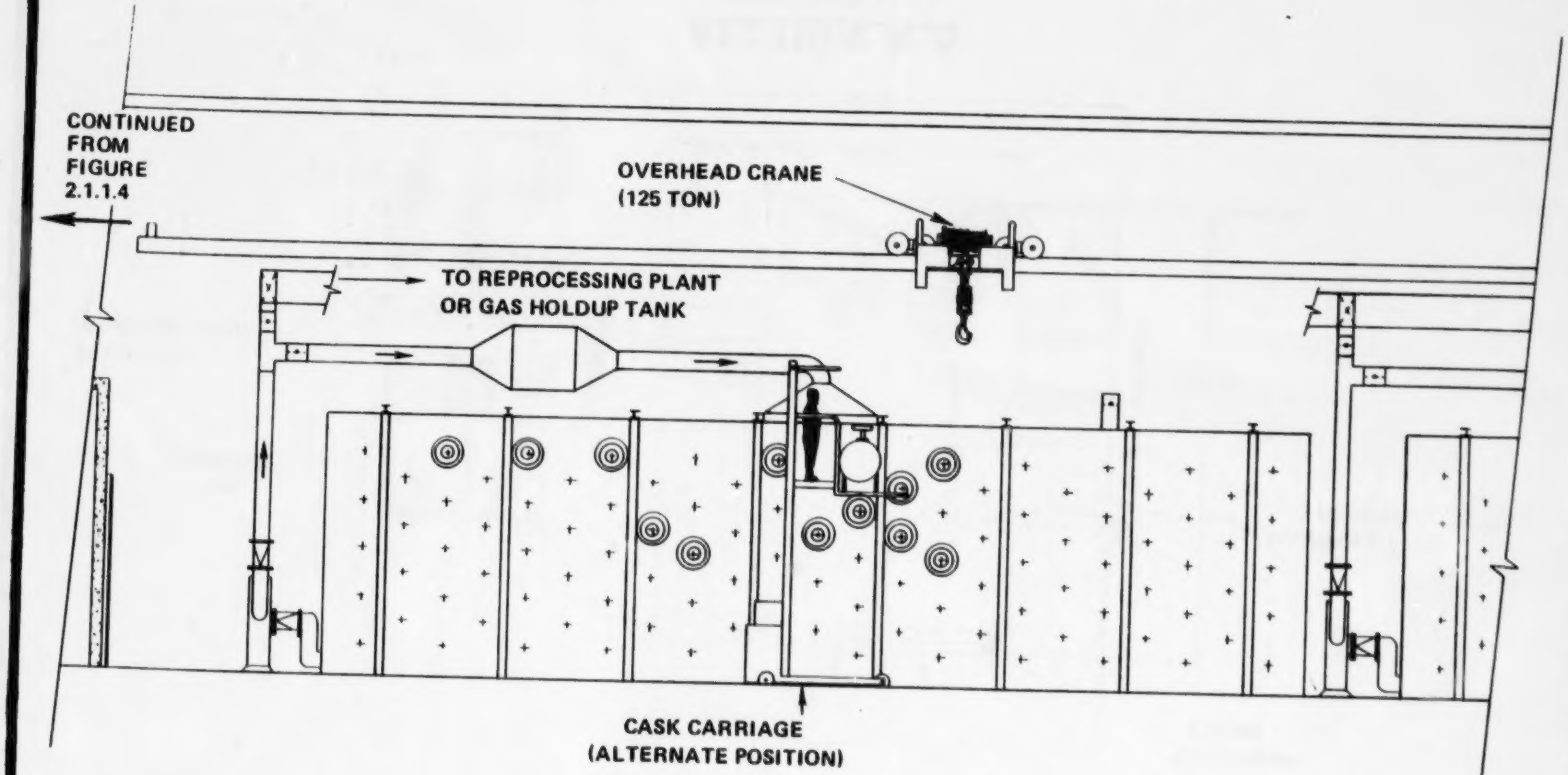

OVERHEAD
CRANE
(20 TON)
HOT CELL
BAY DOOR
CONTINUED
ON FIGURE
2.1.1.5
SHIELDED
TRANSFER
CASK
CARRIAGE
AIR LOCK
FORK LIFT
&
SHIELDED
TRANSFER
CASK
CASK
CARRIAGE
SECTION C-C
FROM FIGURE 2.1.1.2

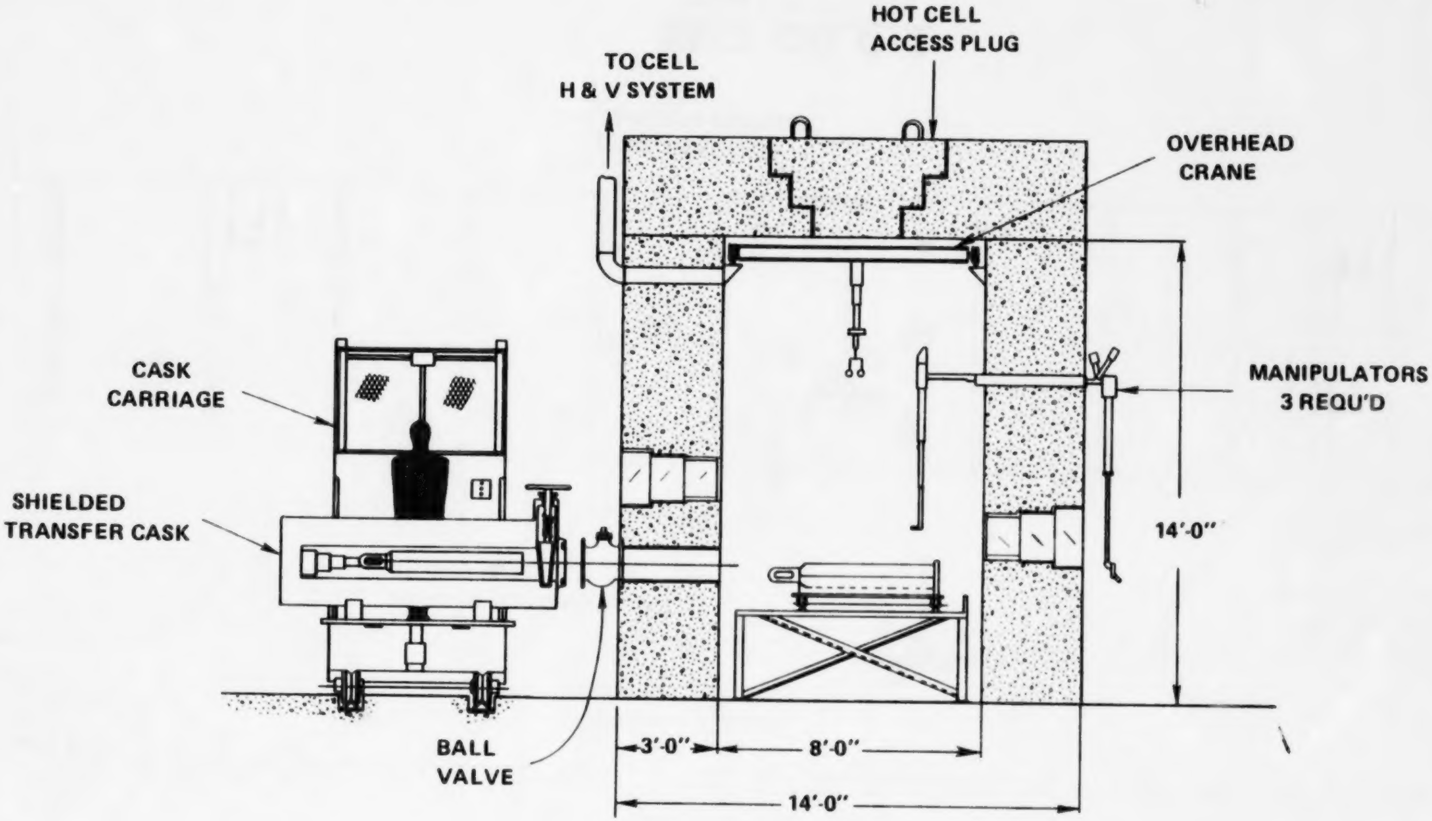
FIGURE 3-4 ELEVATION VIEW OF KSF NEAR HOT CELL
(SOURCE: REF. 3)


CONTINUED
FROM
FIGURE
2.1.1.4
OVERHEAD CRANE
(125 TON)
TO REPROCESSING PLANT
OR GAS HOLDUP TANK
CASK CARRIAGE
(ALTERNATE POSITION)

SECTION C-C

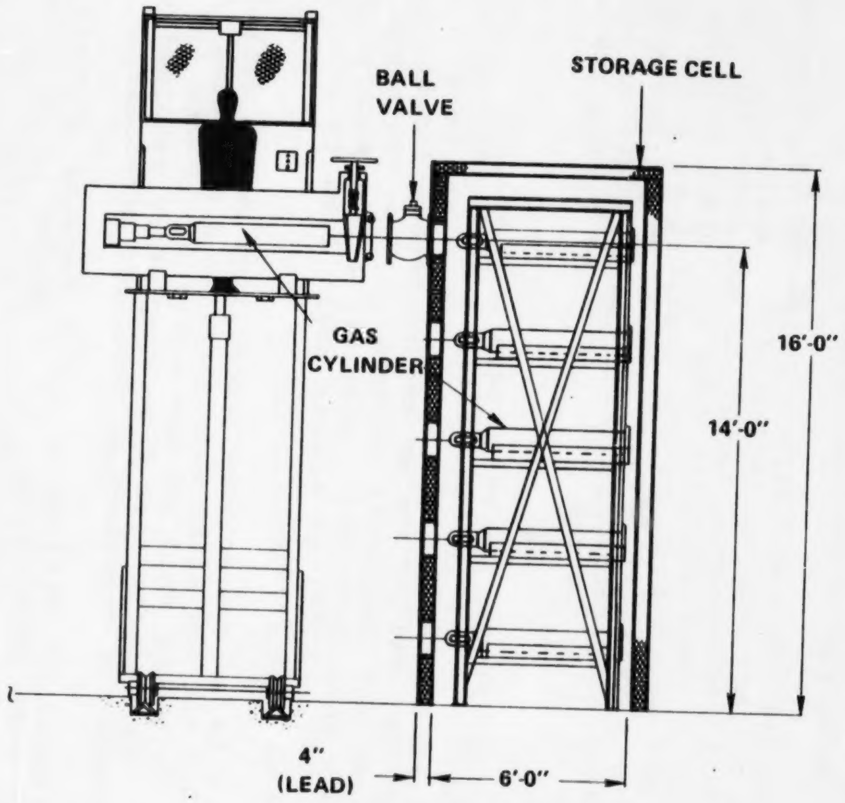
FROM FIGURE 2.1.1.3

FIGURE 3-5 ELEVATION VIEW OF KSF AT STORAGE CELLS
(SOURCE: REF. 3)


TO CELL
H & V SYSTEM
HOT CELL
ACCESS PLUG
OVERHEAD
CRANE
CASK
CARRIAGE
MANIPULATORS
3 REQU'D
SHIELDED
TRANSFER CASK
14'-0"
BALL
VALVE
3'-0"
8'-0"
14'-0"

SECTION A-A
FROM FIGURE 2.1.1.2

FIGURE 3-6 END VIEW OF KSF HOT CELL
(SOURCE: REF. 3)


STORAGE CELL
BALL VALVE
GAS CYLINDER
16'-0"
14'-0"
4" (LEAD)
6'-0"

SECTION B-B

FROM FIGURE 2.1.1.3

FIGURE 3-7 END VIEW OF KSF STORAGE CELL
(SOURCE: REF. 3)

oxygen could lead to serious corrosion in spite of the lessened concentration of krypton-85.

3.1.1.2 Environmental Assessment

The facility is assumed to receive 15,000,000 Ci of radiokrypton per year during its period of loading. Thus, after 30 years of loading the KSF would have 199,000,000 Ci (Figure B-1) distributed among 4,350 cylinders, or an average of about 45,000 Ci per cylinder. The "hottest" cylinder would have 104,000 Ci and the "coolest" would have 15,000 Ci at the end of the loading period. Figure B-1, Appendix B, illustrates the subsequent decay and removal of radioactivity.

Leaking cylinders and valves in the KSF could presumably be handled in the hot cell as indicated in Section 3.1.1.1. There would be some radiological hazard to the fork-lift operator in transferring a leaky cylinder from a storage cell to the hot cell. There would also be the possibility (10^{-1} per year) of dropping a cylinder with a loss of its krypton-85 in the vicinity of the operator.[3] These hazards could be lessened by remote operation as in the DuPont plan or by the use of double containment.

Each cell of the KSF would have to dissipate 17 to 19 kW of decay heat upon the initial filling with 500 psi cylinders. Table 3-1 reproduces Brown's estimates of the conditions within a cell if the cooling system should become inoperative.[3] The temperatures and pressures imposed on the cylinders would be well within specifications. However, a temperature outside the cell of 67°C (153°F) or higher might lead to cell pressures in excess of safe levels.

There is an appreciable probability that a cylinder or its valve would corrode after many years to give sudden failure. A release into the cell of the order of 10^3 to 10^4 Ci could result. The release should be completely retained within the cell and could be recovered as described previously.

Pressurized cylinders would require surveillance and probably some rebottling over a period of 50 to 80 years.[3] Appendix B shows that a facility's release of 0.1 percent per year and the eventual venting of all cylinders through a tall stack would result in the nominal annual amount of radiokrypton exposing the world population to 780,000 Ci. This would result in one-third health effect, on the average.

Severe, improbable threats to radiological safety would be earthquakes and tornadoes if beyond design limits. These could release as much as the contents of the entire KSF structure (as much as 200,000,000 Ci). About a tenth of the latter might be lost by the damage of an airplane crash since it would affect only a few cells. Sabotage could produce the loss of the contents of the whole structure.

TABLE 3-1

STEADY-STATE CONDITIONS IN A KSF CELL FOR 500 PSI CYLINDERS AFTER LOSS OF COOLING*

(Source: Ref. 3)

Outside Cell Temperature (°C)	Cell Air Temperature (°C)	Cell Pressure** (atm(abs))	Cylinder Temperature (°C)	Cylinder Pressure (atm)
27	64	1.15	109	55.1
47	85	1.22	130	58.4
67	106	1.29	150	61.7

*104 cylinders containing radioactive krypton, initially at 500 psi and 60°C.

**Excess pressure may need to be vented if beyond storage cell strength.

144<

3.1.1.3 Estimate of Cost

The cost is considered here for storing a nominal year's loading of radiokrypton, as specified in Section 2.1.4.

The currently available estimates[5-6] for a KSF taking three decades to fill indicate the total capital cost for the first decade of loading to be $75,000,000 (±30%) minus about $7,500,000 assumed for interest during construction or a net of $67,500,000. (Interest during construction is not in the figures for the DuPont study with which this is to be compared.)[4] The net capital cost for each of the next two decades would be $38,700,000 (±30%). These values are in 1976 dollars. The total would be $179,000,000 (1979) at a loading rate of 15×10^6 Ci/yr.

The DuPont study[4] indicates the facility for off-gas treatment will cost 56,000,000 dollars (1978$) at 150 percent of the above reprocessing rate. Close to half of this can be attributed to the reprocessing and storage of krypton-85. The DuPont study is sized for twenty years of loading for storage instead of thirty. Hence one can develop an approximate relation (in 1979 dollars),

$$\$30,000,000 = 1.5r + 0.67s$$

where r and s represent capital costs for reprocessing and storage in the facility studied in this report. The cost to be attributed to the present reprocessing r would be 3,000,000 1979 dollars according to Reference 9 and about 4,000,000 1979 dollars according to Reference 10, but 23,000,000 1979 dollars according to Reference 11. The geometric mean, or 6,500,000 dollars, will be taken for the reprocessing capital cost r. Hence s = 30,000,000 dollars according to these sources. These costs contain no interest during construction or money for contingencies.

The basic capital cost in 1979 dollars to be used for the present storage facility is taken to be the geometric mean of $179,000,000 and $30,000,000, i.e., $73,000,000. The geometric mean is used rather than the average in order not to overemphasize the figure of $179,000,000 which is suspected to be too high. The $73,000,000 includes the architect-engineer fee. This figure will be taken to represent the KSF,[3] which is described in more detail than the DuPont facility.[4] Because of the uncertainties in the figure, it is considered useful only for purposes of comparing cost.

The annual operating costs of the KSF, not including the costs of cylinders or presumably the bottling room, have been given as $900,000 (+50%, -25%) (1976$). This estimate includes maintenance.

On the basis of storing 169 m^3 (STP) of radiokrypton the cost estimate for the KSF is calculated as follows:

	Annual Cost, $K
Contribution of capital cost = (1/30)(73,000)	2,433
KSF operating expenses:	
during loading	1,100
during subsequent periods = (50/30)(1,100)	1,850
Total for KSF exclusive of cylinders and bottling room	5,383
145 cylinders at 0.5 $K each = 145x0.5	73
Bottling room:	
Capital costs:	
Room of 1-1/2 ft concrete + 3 in. Pb = (1/30)(250)	8
Compressor of 10-yr life = (/10) (30)	3
Installation costs (100%) = 8 + 3	11
Personnel costs:	
One operator half-time = (1/2)(40)	20
Maintenance	5
	47
Total for KSF, cylinders and bottling room	5,503

Adding 30 percent for contingencies brings the latter to 7,150 $K (±40%).

3.1.2 Storage in a Dry Well

The use of a dry well at the surface of the earth for the storage of spent fuel is being studied in connection with Spent Fuel Storage and Isolation at the Nevada Test Site and the handling of CANDU fuel elements at Chalk River, Canada.[12] It is also being considered for the storage of 385-liter tanks of approximately 500 psi radioactive krypton in essentially a SURF arrangement (Figure 3-8).[13] (SURF stands for Spent UnReprocessed Fuel which is to be stored in a dry well such as this.) At these pressures the temperature of the tank depends markedly on whether or not the medium is loose soil, basalt, tuff, granite, argillite or sand, etc. The cost of the dry well also depends upon the medium. Although the estimates of cost for this option have not yet been published, they likely will be

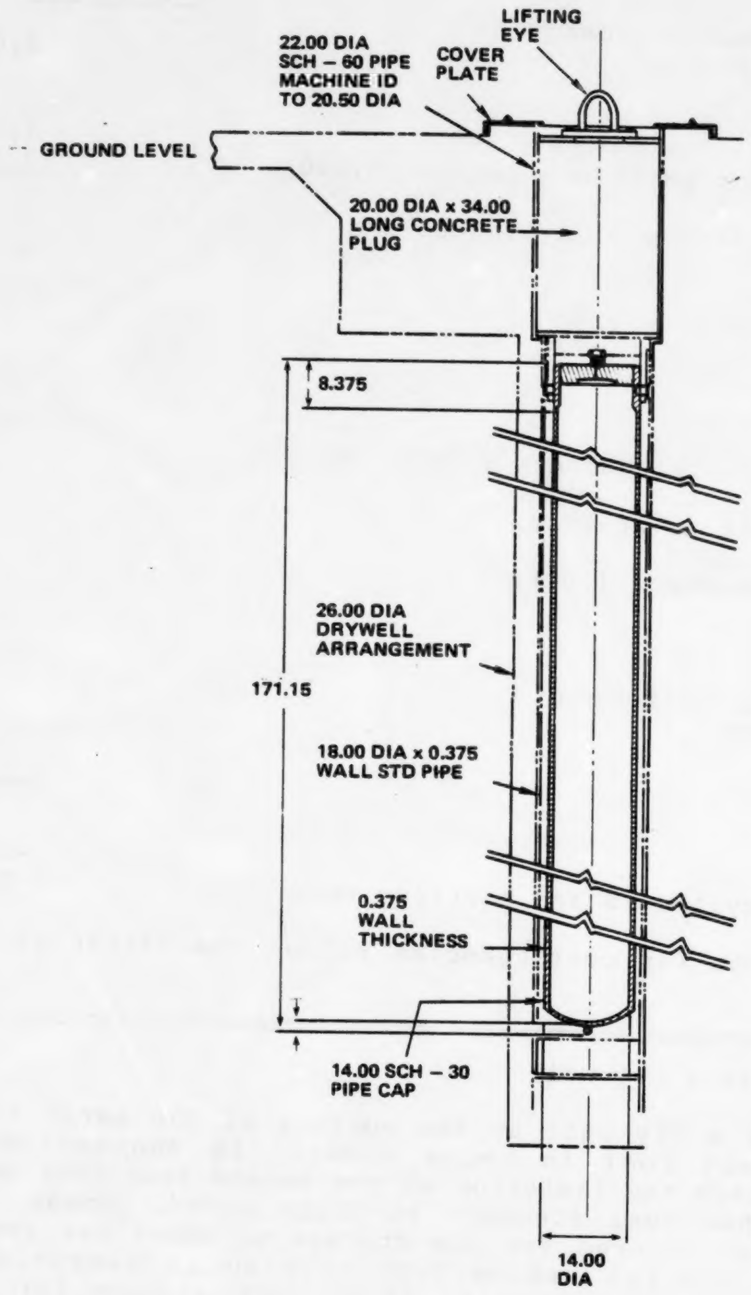

LIFTING EYE
22.00 DIA SCH – 60 PIPE MACHINE ID TO 20.50 DIA
COVER PLATE
GROUND LEVEL
20.00 DIA x 34.00 LONG CONCRETE PLUG
8.375
26.00 DIA DRYWELL ARRANGEMENT
171.15
18.00 DIA x 0.375 WALL STD PIPE
0.375 WALL THICKNESS
14.00 SCH – 30 PIPE CAP
14.00 DIA

FIGURE 3-8 SURF DRY WELL CONFIGURATION[13]

(DIMENSION IN INCHES.)

less than that of the KSF with 500 psi cylinders. However, the KSF would have better containment of the radiokrypton than could be afforded by a 500 psi tank in a SURF dry well.

3.1.3 Storage in a Deep Geological Medium

3.1.3.1 General Aspects

It was stated in Section 2.3 that pressurized gas cylinders have too high a possibility of leakage to be stored without surveillance and the capability for remedial action. For the same reason there must be a leak-proof secondary containment such as a negative-pressure concrete cell anticipated for the Krypton Storage Facility.[3] The secondary container would have to permit the ready detection of leakage of the pressurized container. The concrete cells for the KSF would have this capability. No alternative secondary container has yet been developed. Hence the KSF concrete cells, and essentially the whole Krypton Storage Facility (except the stack), are considered here to be located in the geological caverns. The caverns would effectively replace the KSF buildings but not their cells. This KSF in a cavern would require appreciable personnel and forced air cooling during the life of the facility. A floor area of 1.90 acres would be required for a KSF which would operate for thirty years.[5,6] A granitic medium is assumed. In general, the geological site would be some distance from the reprocessing plant that would produce the krypton-85 and costs for transportation would be incurred.

3.1.3.2 Environmental Assessment

The radiological impact at the facility would be similar to that of an above-ground KSF (Section 3.1.1) except that there would be no vulnerability to tornadoes or crashing aircraft and reduced vulnerability to sabotage and earthquakes. The possibility of water leaking into the caverns would have to be considered, but the concrete cells should protect against that.

3.1.3.3 Estimated Cost

The cost is estimated for storing the nominal annual production of radiokrypton. The cost of the aboveground KSF for 500 psi cylinders (Section 3.1.1.3) is taken minus the cost of the building (estimated at \$50/ft^2). A suitable cost for the geological part is added to this. Use of an abandoned mine or a newly mined facility having a horizontal tunnel are both considered.[14]

	Annual Cost, $K
Total for KSF, cylinders and bottling room (Cf. Section 3.1.1.3)	5,503
10% premium on KSF capital costs for underground construction	243

	Annual Cost, \$K
Credit for unused building = 82,000 ft^2 (0.050 \$K/ft^2)/30	- 138
Total exclusive of geological cost, casks and transportation	5,608

	Abandoned Mine	Horizontal Tunnel
Geological cost:		
From capital costs:	67	111
Engineering (5% of the above)	3	5
	70	116
Costs of 48 casks of Blomeke-Perona type[15] (70 \$K each, 30-yr life, 3 cylinder/cask) = 48 x 70/30	112	112
	182	228
Transportation to:		
Eastern US site	37	37
Western US site	99	99
Total costs:		
Eastern US site	5,827	5,873
Western US site	5,889	5,935
Adding 30% for contingencies brings the latter figures to		
Eastern US site	7,580	7,630
Western US site	7,660	7,720

The error in these estimates is ± 40%.

3.1.4 Storage in the Ocean

3.1.4.1 General Aspects

The 500 psi radiokrypton would be contained in the equivalent of 6K cylinders of 42.5 l of the kind described in Section 2.4. The cylinders would be high-nickel alloy for external corrosion resistance and strength. The valve of the cylinder would be replaced with a more leak-proof, welded closure since retrievablility would not be sought. Each water-filled cask of the type of Blomeke and Perona[5] would contain three cylinders. Hence 48 casks and 29 projectiles would be required annually. The radiokrypton is assumed to be bottled in a bottling facility at the reprocessing plant. The cylinders are then inspected in a hot cell such as that at the KSF, and placed in their casks for transportation, shipping and disposal.

3.1.4.2 Environmental Assessment

The sudden loss of radiokrypton from a cylinder in its cask has been described in Section 2.3. However, once a cylinder is in the seabed, the gradual leakage of its 10^5 Ci would probably lead to a negligible appearance of radioactivity at the surface of the ocean. The radioactivity should dissolve in the abyssal water in the sediment and at the seafloor, and this water should not circulate to the surface of the ocean for centuries. If the krypton were in the form of the hydrate, there would be no gaseous krypton either before or after a sudden, gross failure of a cylinder. Then the krypton would remain as the solid hydrate and dissolved krypton in the sediment. However, if the krypton were not as hydrate but as gas before a sudden gross failure of a cylinder, then bubbles could conceivably move upward a considerable distance before dissolving in the seawater.[16] It is doubtful that the bubbles could reach the surface of the ocean more than two miles above, but some dissolved radioactivity might circulate to the surface.

The emplaced cylinders would not be subject to the effects of tsunamis, hurricanes, tornadoes, plane crashes or sabotage. The cylinders would probably be undamaged by the infrequent earthquakes, but a rare volcanic action could release radioactivity.

3.1.4.3 Estimate of Cost

The cost estimated for storing the nominal annual production of radiokrypton is as follows:

	Annual Costs, $K
Bottling room:	
Capital costs:	
Room of 1-1/2 ft concrete and 3 in. of Pb = (1/30)(250)	8
Compressor of 10-yr life = (1/10)(30)	3
Installation costs (100%) = 8+3	11
Gas bottles with welded closures, each of 350 lbs at 0.020 $K/lb = 145x350x0.020	1,015
	1,037
Personnel in bottling room:	
One operator half time = (1/2)(40)	20
Maintenance of bottling room:	5
Total for bottling and cylinders	1,062

	Annual Costs, $K
Hot cell and loading of shipping casks:	
Capital costs of KSF type hot cell, offices, change rooms, etc., with facility for loading casks (Cf. Section 3.1.1.3) = (1/30)(10,000)	333
Concomitant operating expenses (Cf. Section 3.1.1.3)	500
Costs of 48 casks (70 $K each, 30 year life) = 48 (70/30)	112
Total for land-based hot cell and loading casks	945
Transportation by land from p. 2-13	39
Total costs of land operations	2,046
Costs of marine portion:	
Refitting of ship with navigational equipment = (1/30)(3,650) from p. 2-13	122
Refitting of ship with shielding and equipment for remote control = (1/30)(950) from p. 2-13	32
Projectiles for cylinders = 29x1.3 from p. 2-13	38
Operational personnel, port charges, and ship rental (3 months of year)	750
Supplies and equipment	3
Architect-engineer	45
Total for marine portion	990
Total of land-based and marine portions	3,036

Adding 30% for contingencies makes the total 3,950 $K (±40%).

REFERENCES

1. C.W. Nielsen, "Safety Analysis Report for Packaging Krypton Shipping Container USA/5958/BL (ERDA-ID)," Report ICP-1077, Idaho National Engineering Laboratory, Idaho Falls, Idaho, June 1975.

2. B.A. Foster and D.T. Pence, "An Evaluation of High Pressure Steel Cylinders for Fission-Product Noble Gas Storage," Report ICP-1044, Idaho Operations Office Idaho Falls, Idaho, Feb 1975.

3. R.A. Brown, et al, eds., "Reference Facility Description for the Recovery of Iodine, Carbon and Krypton from Gaseous Wastes," Report ICP-1126, National Technical Information Service, U.S. Dept. of Commerce, Springfield, Va.

4. E.I. DuPont de Nemours & Co., "Design Integration Study, Spent LWR Fuel Recycle Complex, Conceptual Design, Case A-1, Separated Streams," Report DP-CFP-78-121, Savannah River Laboratory, Aiken, S.C., Nov 1978.

5. "Environmental Impact Statement: Management of Commercially Generated Radioactive Waste," Report DOE/EIS-0046-D, Department of Energy, Washington, DC, April 1979.

6. "Technology for Commercial Radioactive Waste Management," Report DOE/ET-0028, Department of Energy, Washington, DC, 1979.

7. T.R. Pinchback and D.A. Knecht, "The Development of Process and Storage Materials Suitable for Krypton-85 Waste Management," paper presented at the International Symposium on Science Underlying Radioactive Waste Management, Boston, Mass., Nov 29-Dec 1, 1978.

8. U.S. Nuclear Regulatory Commission, "Standards for Protection against Radiation," Title 10, Code of Federal Regulations, Part 20, in the Federal Register, Jan 1, 1976.

9. C.M. Slansky, "Nuclear Power Growth Spurs Interest in Fuel Plant Waste," Environ. Sci. Technol. 3 (5) 446 (1969).

10. B.C. Finney, et al, "Correlation of Radioactive Waste Treatment Costs and the Environmental Impact of Waste Effluents in the Nuclear Fuel Cycle--Reprocessing Light-Water Reactor Fuel," Report ORNL/NUREG/TM-6, Oak Ridge National Laboratory, Oak Ridge, TN, Jan 1977.

11. L. Kovach, "Technology and Cost Evaluation of Krypton-85 Control for Nuclear Fuel Reprocessing Plants," Report NUCON 128, prepared by Nuclear Consulting Services, Columbus, Ohio, for U.S. Environmental Protection Agency, Office of Radiation Programs, Washington, DC, Aug 1976.

12. T.D. King, et al, "Near-Surface Heat Sink Storage," pp. 17.40 to 17.47, in J.W. Bartlett, et al, eds., "Alternatives for Managing Wastes from Reactors and Post-Fission Operations in the LWR Fuel Cycle," Report ERDA-76-43, vol. 3, available from the National Technical Information Service, U.S. Dept. of Commerce, Springfield, Va.

13. R.D. Klett, ed., "Krypton-85 Disposal Program, Semiannual Report, Aug 15, 1977 to Mar 31, 1978," Report SAND78-1667, Sandia Laboratories, Albuquerque, N.M., Feb 1979.

14. P. Macbeth, et al, "Evaluation of Alternative Methods for the Disposal of Low-Level Radioactive Wastes," Report NUREG/CR-0680 to the U.S. Nuclear Regulatory Commission, Low-Level Waste Branch, by Ford, Bacon & Davis Utah, Section 3.4 (1979).

15. J.O. Blomeke and J.J. Perona, "Management of Noble-Gas Fission Product Wastes from Reprocessing Spent Fuels," Report ORNL-TM-2677, Oak Ridge National Laboratory, Oak Ridge, TN, Nov 21, 1967.

16. J. Mishima and L.C. Schwendiman, "Some Observations on Noble Gas Partitioning in Dynamic Air-Water System," in M.W. First and J.M. Morgan, Jr., eds., Proceedings of the Eleventh AEC Air Cleaning Conference, Richland, Wash., Aug 31- Sept 3, 1970, Vol. 5, Report CONF 700816-5, U.S. Atomic Energy Commission, Dec 1970.

3.2 STORAGE IN CYLINDERS AT 2000 PSI

The heat output of a 50-l gas cylinder (size T) containing 2000 psi of the radiokrypton would be 611 watts for a wall temperature of 127°C (261°F), according to B.A. Foster and D.T. Pence.[1] The heat output would be even higher if the wall temperature were lower and the pressure were the same.

3.2.1 Storage in a Building

3.2.1.1 General Aspects

The size T cylinder is rated for 2400 psi and if loaded into the Krypton Storage Facility, cylinder pressures of 3200 psi could result if the air cooling failed. It would be beneficial to have an emergency spray cooling sytem, but even this could fail. A remedy might be to use a cylinder of the 6K size (DOT Specification 3AA 6000) which is rated for 6000 psi. It is made of A.I.S.I. 4130 steel. It holds 42.5 l of gas. This cylinder is 1 in. larger than that of the size T cylinder, thus requiring only minor changes in the KSF to accommodate it.[2] It is assumed that a suitable all-metal valve or closure could be developed to handle at least 4000 psi of radiokrypton. It is also assumed that impurities other than noble gases be small.

The method of heat-transfer analysis of R.B. Bird, et al,[3] also used by Foster and Pence,[1] indicates that the 6K cylinder and its 2000 psi gas would generate 660 watts and be at 83°C (181°F) if the ambient air within the cell were at 21°C (70°F). Due to a favorable compressibility,[4] each such cylinder would hold 450,000 Ci so that 33 of them would hold 169 m^3 (STP) of radiokrypton. Only 28 percent as much storage space would be required in a KSF. The air cooling of the KSF cell would have to be increased by a factor of about 4, with duplicate equipment. A backup of spray cooling would be installed as would an additional inch of lead shielding. Most other changes would be minor.

3.2.1.2 Environmental Assessment

Using the same loading schedule as with 500 psi cylinders (Section 3.1.1.2), the KSF would have 990 cylinders of 2000 psi after 30 years of loading and they would range from 450,000 Ci in the "hottest" cylinder to 34,000 Ci in the "coolest." Each cell of the KSF would be filled in 3.15 years at which time the cell's decay heat would be 63 kW (Equation B.2). Table 3-2 contains estimates of the conditions within a cell if the air- and spray-cooling systems became inoperative. Calculations of Reference 2 were extended assuming that thermal conduction was principally through the cell wall. The worst case of a temperature outside the cell of 67°C (153°F) could lead to the gas within the cylinder being at 297°C and 3700 psi. The

TABLE 3-2

STEADY-STATE CONDITIONS IN A KSF CELL FOR 2000 PSI CYLINDERS AND CESSATION OF COOLING*

Outside Cell Temperature (^oC)	Cell Air Temperature** (^oC)	Cell Pressure*** (atm(abs))	Cylinder Temperature (^oC)	Cylinder Pressure (atm)
27	158	1.60	250	229
47	182	1.70	275	243
67	205	1.79	297	253

*104 cylinders containing radioactive krypton, initially at 2000 psi and 83^oC.

**Estimated from Table 3-1 assuming thermal conduction is principally through the cell wall.

***Assuming no venting, but venting would probably be necessary to prevent physical damage to the cell.

6K cylinder would probably withstand these conditions, but experimental verification would be in order.

A gas cylinder of 2000 psi would almost certainly require surveillance unless there is a technical breakthrough in the valve or closure technology. It might be unrealistic to expect either a 2000 psi or 500 psi gas cylinder not to gradually leak over a period of 50 years. The leakage would depend upon the purity of the gas and its pressure for a given type of closure. The leakage could be kept within 0.1 percent per year by satisfactory surveillance, repair, rebottling, etc. It has been shown in Appendix B that 169 m^3 (STP) of radiokrypton in the facility released at 0.1 percent per year from a tall stack would commit the world, on the average, to one-fourth as much radiological exposure as the scheduled release from removed cylinders. The total would give one-third health effect.

The vulnerability of the facility to natural hazards, sabotage, etc., would be similar to the 500-psi case (Section 3.1.1.2), except that each cell would contain four times as much radioactivity.

3.2.1.3 Estimate of Cost

The basis of the 500-psi case (Section 3.1.1.3) is used in estimating costs for storage in 2000 psi cylinders except as indicated. The costs of maintenance and supplies, together with those of power, including those of refrigeration, are assumed to be the same. Only 28 percent as many cells of KSF size would be used, but there would be added expense for a spray-cooling backup and an additional inch of lead in the cells.

	Annual Costs, $K
Capital costs of KSF offices, change rooms, etc., 150 ft stack, dump tank, spare storage tank, and head end (Cf. Section 3.1.1.3 and References 5 and 6) = (1/30) (14,500)	483
Costs of basic KSF storage cells	537
1 in. of extra Pb in cells	19
Spray cooling system	33
Total KSF operating expense (Cf. Section 3.1.1.3)	2,950
Total for KSF exclusive of cylinders and bottling room	4,022
33 cylinders of 6 K type at 0.6 $K each = 33x0.6	20
Bottling room:	
Capital Costs:	
Room of 11/2 ft concrete + 4 in. of Pb = (1/30)(260)	9

	Annual Costs, $K
Compressor of 10-yr life = (1/10) 60	6
Installation costs (100%) = 9+6	15
Personnel costs:	
One operator (one-quarter time) = (1/4)(40)	10
Total for KSF, cylinders and bottling room	4,082

Adding 30 percent for contingencies brings the latter to 5,310 $K (±40%).

3.2.2 Storage in a Dry Well

The 2000 psi radiokrypton could not be contained in a 385-l tank such as R.D. Klett, et al, envision for a dry well of the SURF type (Section 3.1.2)[7] unless the tank were made stronger and improved cooling were applied. The improved cooling might be in the form of forced air, fins or heat pipes, for example. The resulting increase in expense might vitiate a possible advantage of cost of storage in a dry well compared with a building, for example. The subject warrants more study.

3.2.3 Storage in a Deep Geological Medium

3.2.3.1 General Aspects

The basic design for the storage of 2000 psi cylinders in a geological repository is considered to be similar to that for 500 psi cylinders (Section 3.1.3.1). In this case a floor area of 0.66 acre would be required, exclusive of the entrance. The number of Blomeke-Perona shipping casks would be reduced from 45 to 33 for the basis 169 m^3 (STP) of radiokrypton.

3.2.3.2 Environmental Assessment

The higher density of stored radiokrypton would make the environmental danger more serious if both the refrigeration and backup shower should fail. However, the 6,000 psi rating of the gas cylinders should minimize this danger.

3.2.3.3 Estimate of Cost

The cost is estimated for storing the nominal annual amount of radiokrypton at 2000 psi in a deep geological repository. As in Section 3.1.3.3 the cost of the aboveground KSF part is diminished by the cost of its floor area at $\$50/\text{ft}^2$ but a 10 percent premium is paid on KSF capital costs for underground construction.

	Annual Costs, $K
Total for KSF, cylinders and bottling room (from Section 3.2.1.3)	4,022
10% premium on KSF capital costs for underground construction	107
Credit for unused building = 28,900 ft^2(0.050 $K/$ft^2$)÷30	- 48
Total exclusive of geological cost, casks and transportation	4,081
Cost of 33 casks (70$K each, 30 year life)	77

Geological cost:

	Abandoned Mine	Horizontal Tunnel
From capital costs:	59	54
Engineering (5% of above)	3	3
	62	57
Transportation to:		
Eastern US site	27	27
Western US site	73	73
Total costs		
Eastern US site	4,247	4,242
Western US site	4,293	4,288

Adding 30% for contingencies brings the latter figures to:

	Abandoned Mine	Horizontal Tunnel
Eastern US site	5,520	5,510
Western US site	5,580	5,570

where the uncertainty is ±40%.

3.2.4 Storage in the Ocean

3.2.4.1 General Aspects

The use of 2000 psi radiokrypton for ocean disposal would be in many ways similar to the 500 psi case of Section 3.1.4. Some significant differences will be discussed here. There would be only 33 cylinders of 2000 psi and the same number of casks. There would be fewer projectiles (7). This 2000 psi, ocean-disposal option will be shown to be comparatively inexpensive and could better tolerate the presence of unseparated xenon than the other options discussed in this report. The reason is partly that a shipping cask could physically accommodate at least four cylinders rather than just one, and the increase in shipping weight and cost would be quite minor. The increased number of cylinders and projectiles would not increase the total cost more than 25 percent. No increased shielding would be required.

3.2.4.2 Environmental Assessment

The comments in this section and the succeeding one are confined to the case of 2000 psi radiokrypton that is free of appreciable quantities of xenon. The 2000 psi cylinders would probably undergo more fractional leakage, however slight, than 500 psi cylinders until they were placed in the sea in their projectiles. The pressure of the seawater would exceed the internal pressure of the 2000 psi cylinder before the latter fell a mile into the ocean. From there on, the external pressure would be better relieved by 2000 psi gas than 500 psi gas. Chances of rupture of the cylinder would be reduced by the higher gas pressure and any leakage in the sediment would still be inward. It should be remembered, however, that the danger of radiation-induced corrosion within the cylinder would probably be worse at 2000 psi pressure.

3.2.4.3 Estimated Cost

The cost of disposing of 169 m^3 (STP) of radiokrypton at 2000 psi in the ocean is estimated as follows:

	Annual Costs, $K
Bottling room costs from capital costs, personnel, and maintenance (Cf. Section 3.2.1.3)	40
Gas bottles with welded closure, each of 350 lbs at 0.020 $K/lb. 33x350x0.020	231
Total for bottling and cylinders	271
Hot cell and loading of shipping casks:	
Capital costs of KSF-type hot cell, offices, change rooms, etc. with facility for loading casks (Cf. Section 3.1.1.3) (1/30)(10,500)	350
Associated operating expense (Cf. Section 3.1.1.3)	400
Costs of 33 casks (70 $K each, 30 year life) 33(70/30)	77
Total for land-based hot cell and loading casks	827
Transportation to port 33x0.700+7x0.185 from p. 2-13	24
Total to shipping port	1,122

	Annual Costs, $K
Costs of marine portion:	
Refitting of ship with navigational equipment (1/30)(3,650) from p. 2-13	122
Refitting of ship with shielding and equipment for remote control (1/30)(1,050) (p. 2-13)	35
Projectiles for cylinders 7x1.3 (p. 2-13)	9
Operational personnel, port charges, and ship rental (3 months of year)	750
Architect-engineer	45
Total for marine portion	964
Total of land-based and marine portions	2,086

Adding 30% for contingencies makes the total 2,712 $K(±40%).

REFERENCES

1. B.A. Foster and D.T. Pence, "An Evaluation of High Pressure Steel Cylinders for Fission Product Noble Gas Storage," Report ICP-1044, Idaho Operations Office, Idaho Falls, Idaho, Feb 1975.

2. R.A. Brown, et al, "Reference Facility Description for the Recovery of Iodine, Carbon and Krypton from Gaseous Wastes," Report ICP-1126, National Technical Information Service, U.S. Dept. of Commerce, Springfield, VA (Apr 1978).

3. R.B. Bird, et al, "Transport Phenomena," p. 415 (1960).

4. A.B. Christensen, "Physical Properties and Heat Transfer Characteristics of Materials for Krypton-85 Storage," Report ICP-1128, National Technical Information Service, U.S. Dept. of Commerce, Springfield, VA, Sept 1977.

5. "Environmental Impact Statement: Management of Commercially Generated Radioactive Waste," Report DOE/EIS-0046-D, Department of Energy, Washington, DC, April 1979.

6. "Technology for Commercial Radioactive Waste Management," Report DOE/ET-0028, Department of Energy, Washington, DC, 1979.

7. R.D. Klett, ed., "Krypton-85 Disposal Program, Semiannual Report, Aug 15, 1977 to Mar 31, 1978," Report SAND78-1667, Sandia Laboratories, Albuquerque, N.M., Feb 1979.

3.3 STORAGE AS A SOLID ALLOY

Experiments currently being made by G.L. Tingey, et al, indicate that krypton may be stably emplaced in certain alloys at concentrations per volume exceeding that in a 2000 psi gas cylinder.[1-3] The most suitable alloys appear to be amorphous. The impregnated alloy is usually capped with a corrosion-resistant metal like nickel to prevent surface attack.

The engineering feasibility has been estimated by scaling up by factors of 3 to 6 from current laboratory apparatus.[1] A hollow cylinder containing ion implanted/sputtered alloy $Fe_{0.79}Y_{0.12}Kr_{0.09}$ would have a length of 56 cm (22 in.), an outside diameter of 16.8 cm (6.6 in.), and an inside diameter of 14.4 cm (5.7 in.). The density of the alloy has been measured to be about 5.8 g/cm^3 by Tingey, et al, giving 0.70 g $*Kr/cm^3$ of alloy. A hollow cylinder would have 1,680 g (3.70 lb) of *Kr, if the volume of its alloy is 2,395 cm^3. Other weights would be 12.2 kg (26.8 lb) of iron plus yttrium, 0.30 kg (0.66 lb) of a nickel cap, and 16.2 kg (35.6 lb) in the copper pipe on the inside of which the alloy was deposited. Thus the total weight of the cylinder would be about 30 kg or 67 lb.

The hollow cylinder would contain 40,000 Ci at loading. Measurements on the release of krypton from the above alloy indicate that the krypton released in 100 years will not exceed 0.001% as long as the temperature remains below 200°C. More advanced alloys would perform even better.

It is shown in Appendix C that the center of the alloy would be less than 1°C hotter than its surface even if the alloy's thermal conductivity were as small as 10^{-2} W/cm-°K. Hence krypton release due to poor thermal conductivity of the alloy appears unlikely.

Although ion implantation/sputtering has considerable promise in producing a storable form for radiokrypton, it is less developed thus far than the methods of storage as a gas. For example, one amorphous alloy disintegrates to a fine powder during the release of krypton above 700°C. The effects on the deposit of impurities other than noble gases are unknown; nor is the effect of xenon well known. The effects of radiation damage and the daughter product rubidium should be studied. However, as noted in Tingey's paper, the prospects for scale-up to handle 169 m^3 (STP) of radiokrypton per year appear to be good.[1]

If the krypton alloy from ion implantation/sputtering were satisfactorily cooled, it might eventually offer the potential for surveillance-free storage for the century or so of the dangerous life of krypton-85. Cooling probably would not be a serious problem because each of the hollow cylinders would emit a maximum of only 58 watts.

3.3.1 Storage in a Building

3.3.1.1 General Aspects

A hollow cylinder containing the radiokrypton is assumed to be a suitable form for storage. With appropriate modification, the Krypton Storage Facility could provide a satisfactory housing.[5,6] The KSF's capability of surveillance is assumed to be retained unless it is demonstrated to be unnecessary. The original size of the storage cell of the KSF is also assumed to be retained (Figures 3-3, 3-5 and 3-7), but the hollow cylinders would be oriented vertically for better cooling. The entrance port for each hollow cylinder would likely be different from that of the original KSF and entrance might be through the top of the cell. In the latter case, a remotely operated overhead crane might be used for loading and unloading at a storage cell. A lateral spacing of 2 in. between cylinders is assumed. The cell of 62 ft length, 6 ft width, and 16 ft height would then accommodate 86x8x6 = 4,128 cylinders, assuming a 2-1/2 ft gas plenum is left above and below the array of cylinders to attain a uniform flow of air coolant. The basis 169 m^3 (STP) Kr contains 632 kg *Kr and would require 632/1.68 = 376 of these cylinders. Thus, 11.0 years would be required to fill one KSF cell at 169 m^3 (STP) Kr per year.

The maximum heat dissipation per cell would be 171 kW. The cooling requirements can be estimated by well-known techniques.[7,8] It is calculated that the maximum temperature in a cylinder would be less than 17°C (30°F) higher than the air inlet temperature for a cell if the flow velocity through the cell were 5 ft/sec. The cooling requirements for a cell would be 640,000 BTU/hr with an air flow of 290 tons/hr. This cooling would be arranged essentially as in the KSF design,[4-6] except that air ducts to the upper plenum of the cell would be from the side rather than the top to prevent interference with the top loading and unloading of cylinders. The refrigeration unit for each cell would be supplied in duplicate. In addition, emergency cooling would be provided in the form of a water spray.

The prospects for retrieving the stored krypton and eventually decommissioning the facility would be very good barring unforeseen circumstances.

3.3.1.2 Environmental Assessment

As with 500 psi gas cylinders, the facility is assumed to receive 15,000,000 Ci per year and would have 199,000,000 Ci after thirty years of loading. The average hollow cylinder with deposited krypton would then have 18,000 Ci, the "hottest" would have 40,000 Ci, and the "coolest" would have 5,800 Ci.

Fortunately the deposited krypton of a hollow cylinder would not present the leakage hazard and expense of a valve on a gas cylinder. If leakage should be detected in a storage cell, each of the hollow cylinders could be transferred to the hot cell for inspection and suitable action using a remotely controlled overhead crane. A cylinder's stored krypton should ordinarily be unaffected by dropping the cylinder or any other mechanical shock.

The most likely cause of leaking krypton would probably be insufficient cooling of the hollow cylinders. A cylinder in transit between the implantation facility and the hot cell or the hot cell and a storage cell would be adequately relieved of its maximum of 58 W by the closely fitting lead shielding. However, there is a possibility that a cell's duplicate refrigeration system and emergency spray cooling might fail even though the plant would have emergency diesel power. If the worst-case thermal conditions for a cell recently filled and dissipating 209 kW are assumed, the temperatures and pressures shown in Table 3-3 occur. The figures in columns 2 and 4 are approximate, but they suggest a possible loss of 1 percent of the radiokrypton in an hour's time when combined with Tingey's data.[1] This lost krypton could be recaptured and reprocessed, or vented through the stack. Experimental work would be required to show whether the nickel cap might be ruptured by this loss of gas. An opening in the cap would expose the alloy to attack by air with a resultant loss of krypton.

A severe earthquake, tornado, airplane crash or instance of sabotage might rupture the protective nickel cap of some of the krypton alloy. This could lead to a gradual loss of krypton. A more serious consequence would probably be a loss of cooling, with consequences suggested by the preceding paragraph. However, the instantaneous release of large amounts of krypton would be unlikely.

3.3.1.3 Estimated Cost

The cost estimates below are based on 169 m^3 (STP) of radiokrypton. G.L. Tingey, et al, have estimated costs for the process of ion implantation/sputtering. Scaling to the present volume and subtracting interest charges, their costs are estimated to be 529 $K from capital equipment, 227 $K for electric power, and 785 $K for materials and fabrication, or a total of 1,541 $K. Designating the latter as "implantation costs," it is necessary to also add costs for the building.

	Annual Costs, $K
Implantation costs	1,541
Building:	
From capital cost:	
(1/30)(1,000)	33

TABLE 3-3

STEADY-STATE CONDITIONS IN A CELL WITH IMPLANTED RADIOKRYPTON AND CESSATION OF COOLING

Outside Cell Temperature (°C)	Cell Air Temperature* (°C)	Cell Pressure** (atm(abs))	Temperature of Krypton Alloy (°C)
27	505	2.6	>505
47	538	2.7	>538
67	571	2.8	>571

*Estimated from Table 3-1 assuming thermal conduction is principally through the cell wall.

**Calculated for no venting, but venting would be necessary to prevent physical damage to the cell.

165<

	Annual Costs, $K
Installation (50%) 0.5(33)	17
Maintenance	10
	60
Total for prior bottling (Section 3.1.1.3)	49
Total exclusive of KSF	1,650
Capital costs of KSF offices, change rooms, etc., 150 ft stack, dump tank, spare storage tank, and head end (Cf. Section 3.1.1.3 and References 5 and 6) (1/30)(14,500)	483
Costs of basic KSF storage cells	127
1 in. of extra lead in cells	4
Refrigeration of 640,000 BTU/hr and 290 tons/hr per cell, 20-year life, for 30 years:	
Redundant units = (1 +30/20)(103)/11.0	23
Power costs (Appendix B) during loading (assuming c=0)	140
Shower backup system	5
KSF operating expense during loading exclusive of refrigeration	987
KSF costs during loading	1,764
KSF subsequent to loading:	
Refrigeration replacement cost (50/20)(103.)/14.2	23
Power costs (Appendix B) subsequently (assuming c=0)	105
KSF operating expense subsequent to loading exclusive of refrigeration (50/30)(987)	1,645
KSF costs subsequent to loading	1,768

Hence, the total for ion implantation/sputtering, bottling room and operation of the KSF would be 5,182 $K. Adding 30% for contingencies brings the total cost to 6,740 $K (±40%).

3.3.2 Storage in a Dry Well

If it is proven that the krypton alloy can be stored without surveillance, then its storage in a dry well could well be satisfactory as well as less expensive than storage in a building. One prerequisite would be adequate cooling. This should preferrably be of the passive type and keep the alloy below some fixed temperature such as 200°C depending upon the particular alloy.

R.D. Klett, et al, are studying the general problem of using a SURF canister[9] (see Figure 3-8). Forty-nine of the aforementioned hollow cylinders could be placed in the SURF canister for initial values of 2.65 kW and 1,800,000 Ci per canister. The matter of heat removal by conduction through the soil deserves special attention.

Storage in a dry well might offer less ready retrieval than storage in a building.

3.3.3 Storage in a Deep Geological Medium

3.3.3.1 General Aspects

The krypton-implanted solid would not depend upon a valve or other closure to prevent the escape of gas. If the alloy were protected by a cap or protective coating, then its cumulative release of krypton-85 should never exceed 0.001 percent for a temperature of 200°C or less, on the basis of non-radioactive tests with the alloy $Fe_{0.79}\ Y_{0.12}\ Kr_{0.09}$. More advanced alloys should leak even less. If tests prove that this degree of tightness persists in the presence of radiation, then this mode of immobilization might not require surveillance or remedial action. This will be the assumption in succeeding analyses, with the implicit assumption that the temperature not rise above about 200°C for this alloy.

Any attack on the alloy beneath its protective cap would release some radiokrypton. Hence, a cap of pore-free nickel is assumed for protection against air, moisture, and particularly salt. Salt could produce hydrogen chloride from the hydrolysis of magnesium chloride to the extent that the latter is present.[10] It is doubtful that the application of an asphaltic, plastic or non-metallic coating over the cap would be desirable because of the impairment of heat transfer and a lack of resistance to radiation.

The hollow cylinders are considered to be mounted on sturdy racks in the cavern's air space.[11] The cooling would be by natural convection. The thermal loading over the room area would be 50 kW/acre produced by a uniform 862 hollow cyliners per acre. This corresponds to 0.44 acre for 169 m^3 (STP) of radiokrypton. If necessary, it would be possible to outfit a cavern with cooling coils in the air space. The use of the coils could be discontinued after the cavern's instrumentation showed a sufficiently low temperature. However, this possibility is not included in the estimate of cost found in Section 3.3.3.3.

3.3.3.2 Environmental Assessment

The hollow cylinders would be remotely loaded onto their racks. A filled room would be sealed after filling in order to passively exclude personnel and provide another barrier against any leaked radioactivity. Any leakage that might occur from stored cylinders would be gently aspirated to a tall stack above ground. Retrieval would be easy using the remote loading apparatus. Eventual decommissioning would not be difficult.

3.3.3.3 Estimated Cost

The cost is estimated for storing 169 m^3 (STP) of radiokrypton. Compare with Section 3.3.1.3. The fourth item below is the cost of a facility for transferring a hollow cylinder from a Blomeke-Perona shipping cask to the loading mechanism.

	Annual Costs, $K
Implantation and prior bottling	1,650
Loading hollow cylinders into casks	15
From capital costs of KSF-type offices, change rooms, etc., above-ground 150 ft stack, and head end Cf. Section 3.1.1.3 and References 5 and 6): (1/30)(14,500)	483
10% premium on KSF capital costs for underground construction	48
From capital cost of loading mechanism:	300
Operating costs	1,645
33 casks (70 $K each, 30-yr life) 33(70/30)	77
Total, exclusive of geological costs and transportation	4,218

Geological cost:[11]

	Abandoned Mine	Horizontal Tunnel
From capital cost:	65	130
Engineering (5% of latter)	3	7
	68	137
Transportation to		
Eastern US site	34	34
Western US site	90	90
Total costs		
Eastern US site	4,320	4,389
Western US site	4,376	4,445
Adding 30% for contingencies brings the latter to		
Eastern US site	5,620	5,710
Western US site	5,690	5,780

where the uncertainty is ±40%.

3.3.4 Storage in the Ocean

3.3.4.1 General Aspects

The hollow cylinders containing radiokrypton could probably withstand the sea if they were plated, coated or clad with nickel. There is no evident requirement for a particular orientation of the cylinder at the bottom of the ocean.[11] Hence, the clad cylinders might be simply dropped off the ship and allowed to sink to the ocean floor. However, implantation within the sediment appears worth the small added expense to gain the extra barrier of stagnant seawater in the sediment. There would be very good passive cooling of a hollow cylinder in any immersed location.

The projectile in Figure 2-3 is assumed to be enlarged 0.10 m in inside and outside diamters to accommodate 4 hollow cylinders abreast in 16 tiers, or 64 cylinders per projectile. Each projectile would dissipate 3.7 kW of heat. Six projectiles and about 33 shipping casks (Figure 3-1) would be required.

3.3.4.2 Environmental Assessment

There would be no danger of the sudden collapse of a container as with gas cylinders, but seawater might penetrate through any pores in the protective nickel exterior. How rapidly this water would attack the underlying alloy is unknown, but it would probably be gradual--i.e., of the order of months or years rather seconds or minutes, depending on the alloy. The gently evolving krypton gas might have time to form stable solid krypton hydrate. In any event, any krypton gas would probably not rise far before dissolving in the seawater. Its circulation to the ocean's surface would probably be slight, if any.

3.3.4.3 Estimated Cost

The cost for storing 169 m^3 (STP) of radiokrypton is estimated as follows:

	Annual Costs, $K
Total implantation and prior bottling costs (Section 3.3.1.3)	1,650
Cost of 1/8-in. Ni coating on hollow cylinders	500
Hot cell and loading of shipping casks:	
Capital costs of KSF-type hot cell, offices, change room, etc., with facility for loading casks (Section 3.2.4.3) (1/30)(10,500)	350

	Annual Costs, $K
Associated operating expense (Section 3.2.4.3)	400
Costs of 33 casks (70 $K each, 30-yr life) 33(70/30)	77
Total for land-based hot cell and loading casks	827
Transportation by land 33x0.700+6x0.185 from p. 2-13	24
Total on land cost of land operations	3,001
Cost of marine portion:	
Refitting of ship with navigational equipment (1/30)(3,650) from p. 2-13	122
Refitting of ship with shielding and equipment for remote control (1/30)(1,050) (Cf. p. 2-13)	35
Projectiles for hollow cylinders 6x1.4 (Cf. p. 2-13)	8
Operational personnel, port charges, and ship rental (3 months of year)	750
Supplies and equipment	3
Architect engineer	45
Total for marine portion	963
Total of land-based and marine portions	3,964

Adding 30% for contingencies gives a total of 5,153 $K (±40%).

REFERENCES

1. G.L. Tingey, et al, "Entrapment of Krypton in Sputter Deposited Metals - A Storage Medium for Radioactive Gases," Report PNL-2879, Battelle Pacific Northwest Laboratory, Richland, Wash., 1979.

2. M.A. Bayne et al, "Krypton Entrapment in Pulse-Biased Sputter-Deposited Metals," Thin Solid Films, 54, 327 (1978).

3. M.A. Bayne, et al, "Krypton Entrapment in Continuous Biased Sputter-Deposited Glassy Metals," presented at the 6th International Vacuum Metallurgical Conference San Diego, CA, April 23-27, 1979, Report PNL-SA-7274, Pacific Northwest Laboratories, Richland, WA.

4. R.A. Brown, et al, eds., "Reference Facility Description for the Recovery of Iodine, Carbon and Krypton from Gaseous Wastes," Report ICP-1126, Idaho Operations Office, Idaho Falls, Idaho, Apr 1978.

5. "Environmental Impact Statement: Management of Commercially Generated Radioactive Waste," Report DOE/EIS-0046-D, Department of Energy, Washington, DC, April 1979.

6. "Technology for Commercial Radioactive Waste Management," Report DOE/ET-0028, Department of Energy, Washington, DC, 1979.

7. J.G. Knudsen, et al, "Heat Transmission," Section 10 of R.H. Perry and C.H. Chilton, eds., Chemical Engineer's Handbook, 5th ed., McGraw-Hill Book Co., N.Y., 1973.

8. B. Gebhart, "Heat Transfer," 2nd ed., McGraw-Hill Book Co., N.Y., 1971.

9. R.D. Klett, ed., "Krypton-85 Disposal Program, Semiannual Report, Aug 15, 1977 to Mar 31, 1978," Report SAND78-1667, National Technical Information Service, U.S. Dept. of Commerce, Springfield, VA, Feb 1979.

10. C.M. Koplik, et al, "Information Base for Waste Repository Design," Report NUREG/CR-0495, Volume 3, Waste/Rock Interactions, prepared for U.S. Nuclear Regulatory Commission, by The Analytic Sciences Corporation, Reading, MA, Mar 1979.

11. P. Macbeth, et al, "Evaluations of Alternative Methods for the Disposal of Low-Level Radioactive Wastes," Report NUREG/CR-0680, prepared for the U.S. Nuclear Regulatory Commission, Low-Level Waste Branch, by Ford, Bacon & Davis Utah, Salt Lake City, UT, April 1979.

3.4 SUBATMOSPHERIC STORAGE

The many years of favorable experience of the Operations Division of Oak Ridge National Laboratory in handling krypton-85 in tanks at less than atmospheric pressure lends support to the technical feasibility of this alternative. One of the main questions, however, is that of cost. In this section atmospheric pressure is taken to be local atmospheric pressure--i.e. the atmospheric pressure at the elevation of the site. This is assumed to be one standard atmosphere unless otherwise stated.

3.4.1 Storage in a Building

3.4.1.1 General Aspects

The building for storage would have some features in common with the envisaged Krypton Storage Facility for 500 psi cylinders.[1] Both cases would have a capability for recovering leaked gas or readily venting it to a stack. Subatmospheric storage would offer retrievability through evacuation of the tanks. Decommissioning could be accomplished with the evacuated tanks.

The storage tanks might be similar to the commercially available FT type of gas bottle. It has a volume of 0.762 m^3 (26.9 ft^3) and weighs 1200 lbs. It ordinarily is made of carbon steel but could be made of other steels, if desired. It would have DOT Specification 106A500X or 110A500W. The FT bottle has an average wall thickness of more than 3/8 in., is rated for a 500-psi working pressure, and could readily withstand a vacuum. Figure 3-9 depicts 48 of these tanks securely mounted in a storage cell. The tanks are outfitted and mounted in the cell before any krypton-85 is stored there. A short length of weldable stainless steel tube is welded to each tank and extends through the adjacent cell wall to a space between the cells. There the tube is field-welded to a length of the same tubing that is welded to a valve at the operator's corridor. The valve should be all metal, perhaps of the 303 SS/monel type.[2] Prior to installation in the cell, each of these tanks and short tubes would have been vacuum heat-treated both to anneal the weld and to outgas the assembly. After it is mounted and field-welded to its mating tube, the assembly would be stored under a vacuum until a final evacuation and filling with the radiokrypton to about three-fourths atmosphere (absolute). Then the valve would be closed. The tubing would also be crimped shut and welded shut on either side of the valve in order to obtain a more leak-proof seal.

The temperature of the gas within each tank would rise but little with respect to the air within the cell because of the low heat flux from the dilute gas. For example, the method of heat-transfer analysis of R.B. Bird, et al,[3] also used by B.A. Foster and D.J. Pence,[4] shows that the tank and gas would be within 5°C of the ambient air within the cell. The gas within

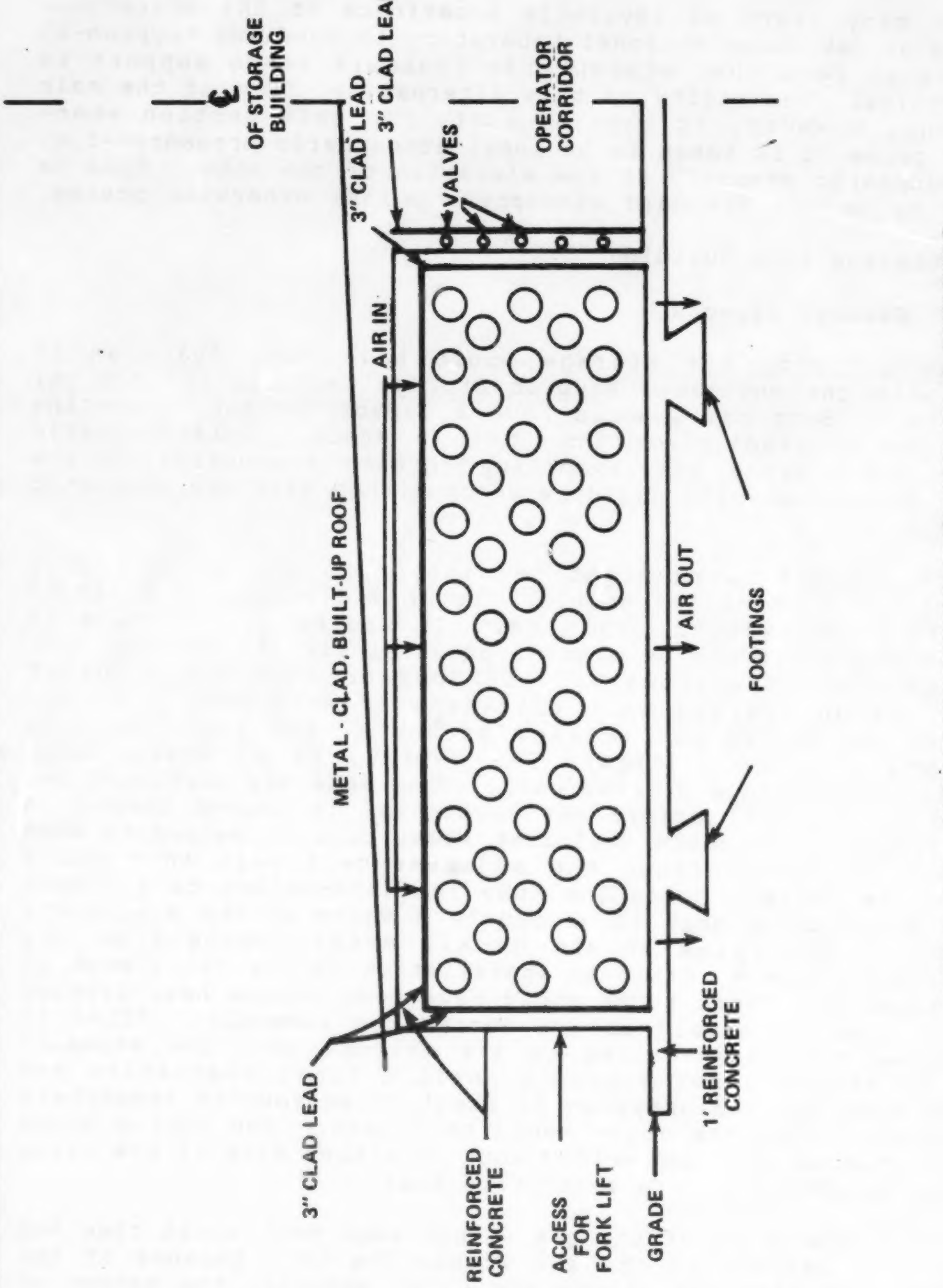

℄ OF STORAGE BUILDING
3" CLAD LEAD
3" CLAD LEAD
VALVES
OPERATOR CORRIDOR
AIR IN
METAL - CLAD, BUILT-UP ROOF
AIR OUT
FOOTINGS
3" CLAD LEAD
1' REINFORCED CONCRETE
ACCESS FOR FORK LIFT
GRADE
1' REINFORCED CONCRETE

FIGURE 3-9 SUBATMOSPHERIC STORAGE IN A BUILDING

173<

the tank could readily be kept below 35°C, thus avoiding the liquefaction of the daughter product rubidium and possibly lessening corrosive effects. The refrigeration system of each cell would be of the closed-cycle type and would be located adjacent to the cell. In the unlikely event of leakage of significant krypton from a tank, the cell's air could be flushed to a nearby holdup tank or a 150-ft stack as in the KSF.[1] The leaking tank could probably be identified by observing its rise in pressure. Then its contents could be pumped out for recovery via its line and valve.

The line of each tank would be connected via its valve to a trunk line behind the lead in the corridor (see Figure 3-9). A trunk line's connections are diagrammed in Figure 3-10. Transferring krypton via a line such as this is more practical than transferring a large, heavy tank such as the storage tank of this concept. The trunk lines would be of a weldable stainless steel. The retrieval of the contents of a tank might be attained with a fusible plug parallel with the tank's valve. The vacuum units symbolized in Figure 3-10 could be a combination of a Roots pump, a throttle valve, and possibly a diffusion pump.[5,6] An alternative to filling the tank via the trunk line would be to lead the tank's valved line to a shielded loading station where the transfer could be made from a pressurized tank. Then the line would be crimped and welded shut on the tank side of the valve.

Corrosive effects should be lower in subatmospheric storage than they are at 500 psi because of lower gas pressure, radiation intensity and temperature. Nevertheless the corrosion tests prescribed for 500 psi gas in a cylinder would be useful here also.[7] Non-inert-gas impurities of the krypton should be below 0.1 percent in the stored gas in order to reduce radiation-enhanced corrosion.[8]

The storage building would have seismic rollers under the footings and be of Seismic Category 1 according to the U.S. Nuclear Regulatory Commission.[9] The Krypton Storage Facility's hot cell, shielded transfer cask, cask carriage, ball valves and associated equipment would not be necessary. Overhead cranes would also be unnecessary. The access for a fork lift (Figure 3-9) would suffice for the initial outfitting of the cell and its eventual decommissioning. As in the original KSF, radiation levels would be reduced by lead so as not to exceed the limits for operating personnel as given by 10 CFR 20.[10] The cooling system for each cell would be small (about 2 ton/hour) and would be provided in duplicate. It would have a radiation monitor which would automatically signal significant release of radioactivity within a cell, so that corrective action could be taken. All areas used by personnel would be suitably monitored with alarm instruments.

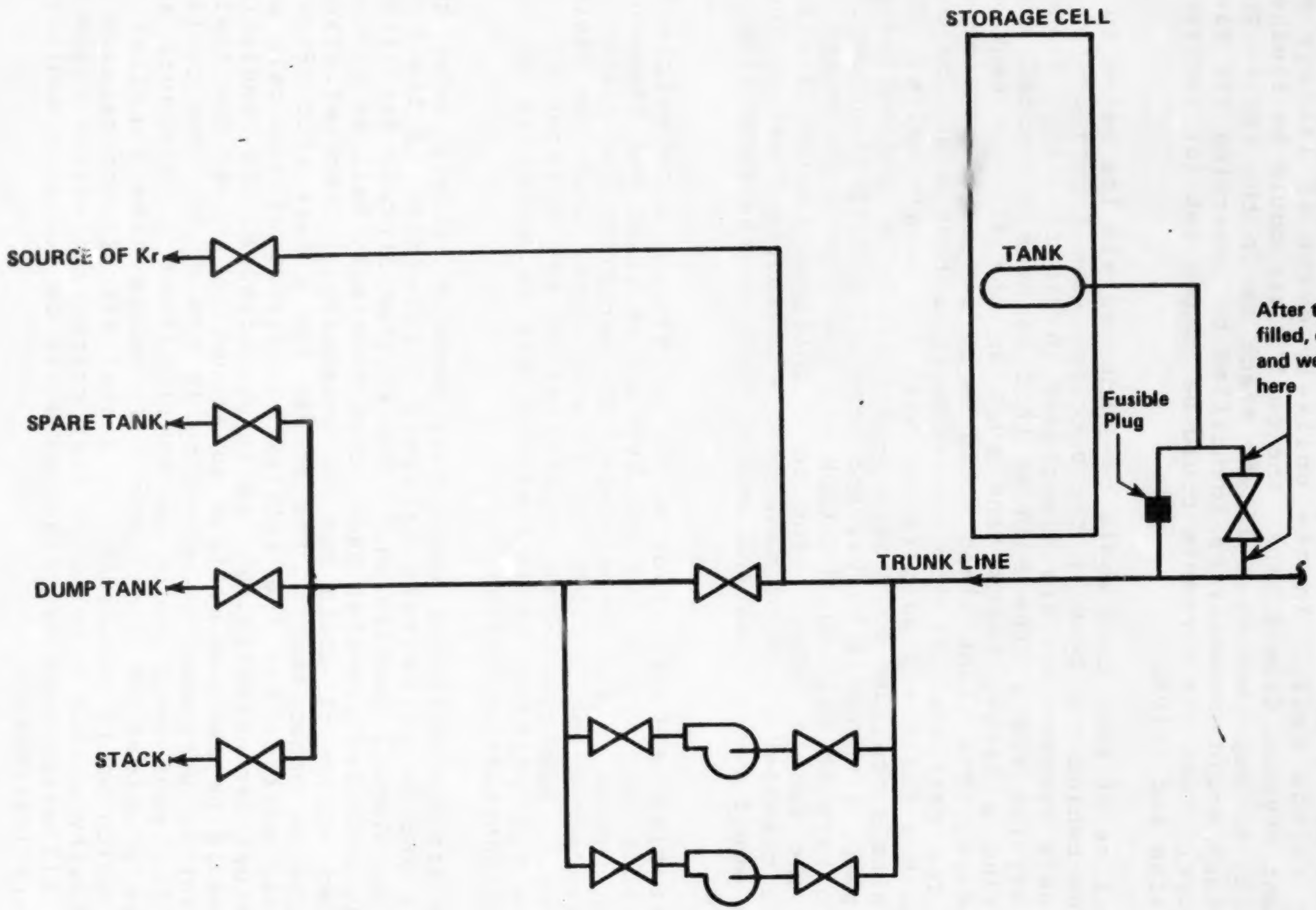

STORAGE CELL
TANK
After tank is filled, crimp and weld shut here
Fusible Plug
TRUNK LINE
SOURCE OF Kr
SPARE TANK
DUMP TANK
STACK

FIGURE 3-10 A TRUNK LINE FOR SUBATMOSPHERIC STORAGE IN A BUILDING

3.4.1.2 Environmental Assessment

The valve bank at the operator corridor would be a potential source of leaked krypton even though each line could be welded to its valve for leak-tightness. Hence the valve area should be under negative pressure with respect to the corridor and a flow of air past the valves should be exhausted to the stack. A badly leaking valve could be detected by aspirating adjacent air to a detector. The valve then could be inactivated by crimping shut the lines adjacent to the valve.

Because of the low gas pressure, each cell in this subatmospheric storage would evolve only 3 kW of decay heat (from 2x10^{6} Ci) when initially filled. This compares with 16 kW (from 11x10^{6} Ci) per cell in the KSF with 500 psi cylinders. The plant would have a diesel generating unit for emergency power. Table 3-4 shows calculated conditions within a cell in the event that its redundant cooling system becomes inoperative. Even for a temperature outside the cell of 67°C (153°F) a subatmospheric pressure would remain within a tank, mitigating against the tank's leakage. Tank temperatures would be only slightly higher than those outside the cell. If the cell should not be vented, the differential pressures across the cell wall would be less than half those for 500 psi cylinders in the KSF (Table 3-1).[11]

The sudden loss of the contents of a tank outside of a cell would be only a remote possibility; only the spare storage tank and dump tank would be outside a cell and they would be filled at subatmoshpheric pressures. These tanks, monitored like the others, could be valved to each other and the stack with suitable pumps. The loss of the contents of a tank within a cell would be less probable than corresponding losses from 500 psi or 2000 psi cylinders in the KSF[1] because of the aforementioned differences in the pressure of impurities, radiation intensity and temperatures.

A severe accident scenario is the possible collapse of the mechanical support for the tanks in a cell due to an earthquake beyond design basis, a tornado, an airplane crash or sabotage. The maximum potential release per cell from one of these improbable events would be 2x10^{6} Ci; the whole facility, filled for 30 years at 15x10^{6} Ci/yr, could release as much as 200x10^{6} Ci (Figure A-1).

3.4.1.3 Estimated Cost

The cost for storing the nominal annual amount of radiokrypton is estimated as follows:

	Annual Costs, $K
Capital costs of offices, change rooms, etc., 150-ft stack, dump tank, spare storage tank, etc.,	

TABLE 3-4

STEADY-STATE CELL CONDITIONS FOR SUBATMOSPHERIC STORAGE AFTER LOSS OF COOLANT

Outside Cell Temperature (°C)	Cell Air Temperature[a] (°C)	Cell Pressure[b] (atm(abs))	Tank Temperature (°C)	Tank Pressure[c] (atm)
27	33	1.03	38	0.79
47	53	1.09	58	0.83
67	73	1.16	78	0.88

[a] Assuming thermal conduction is principally through the cell wall.

[b] Assuming the cell is not vented.

[c] Assuming the tank to have been initially filled at 21°C and 0.75 atm (abs).

	Annual Costs, $K
from R.A. Brown[1] and Section 3.1.1.3 (1/30)(5,200)	173
Storage area:	
Concrete	199
Roof (metal cladding, build-up roofing, insulation and trussing)	21
Refrigeration for 30 years	
Redundant units of 10-year life	38
Power consumption (Appendix B) assuming c=0:	55
Maintenance	13
296 tanks at 1.4 $K each	414
Installation (100%)	414
Lead clad with 10 gage SS on one side and 10 gage carbon steel on the other[1]	660
Installation (50%)	330
Subtotal	2,317
Valves at 0.15 $K each, one per tank 296x1x0.15	44
316SS, seamless tubing	25
Installation (100%)	25
Vacuum pumps of 10 yr life, one for each trunk 10/10	1
Installation (100%)	1
Pressure gage for each tank 296x0.020 $K	6
Installation (100%)	6
Monitoring with alarms (3$K per cell)	18
Installation (100%)	18
Subtotal	144
Capital costs for loading	2,461
Operating costs (cf. Section 3.1.1.3)	
- During loading	900
- During storage	1,500
	2,400
Refrigeration subsequent to loading period	
Redundant units of 10 yr life	60
Power consumption (Appendix B)	41
	101
Total cost	4,962

Adding 30 percent for contingencies brings the latter to 6,450 $K (±40%).

3.4.2 Storage in a Dry Well

The use of a dry well at the surface has been briefly described for 500 psi gas in Section 3.1.2. The subatmospheric storage in a dry well could use a large tank such as the FT type of 30 in. diameter and 82 in. length. Storage would be in a suitably sized dry well of the SURF type, slightly modified. The annular space about the tank in the dry well would be gently aspirated to a 150 ft stack. The aspiration line would be monitored so that if the tank should leak, the gas in the annulus could be flushed to a nearby holdup tank. The containment capabilities would be similar to those described in Section 3.4.1. The initial outfitting, retrieval and eventual decommissioning also would be analogous.

All surface lines and all valves would have 3 in. or 4 in. of clad lead shielding as in Section 3.4.1. These lines and valves also would be monitored as described in Section 3.4.1. Because each 0.75-atm tank would generate only 62 W upon filling, the heat would be dissipated to the atmosphere with only a small temperature elevation of the tank with respect to the medium.[12] The medium is assumed to be soil of average thermal conductivity. Environmental impact could be more favorable for subatmospheric storage in a dry well than in a building because the dry well has infallible passive cooling and is less vulnerability to a tornado or airplane crash. However, the possibility of flooding would need to be minimized.

The following estimate of cost is made with the cost of a dry well represented by DW$, i.e., number of dollars per dry well. The value of a dry well as a trade-off will thus be estimated. No shielded loading vehicle would be required because the radiokrypton would be loaded and retrieved through a transfer line. The dry wells are assumed to be arranged on 5 ft centers, twenty deep, on either side of a shielded trunk-line corridor to accommodate 10 years of loading. Grout poured about the outer container of the dry well should protect it from soil corrosion. The dry wells are assumed to be on the site of the reprocessing plant but transportation costs must be added to the estimate if not. On the same basis as Section 3.4.1:

	Annual Costs, $K
Capital costs of offices, change rooms, etc., 150 ft stack, dump tank, spare storage tank, etc., from R.A. Brown[1] and Section 3.1.1.3 (1/30)(5,200)	173
Storage area:	
296 dry wells	0.296(DW$)
296 tanks at 1.4$K each	414
Installation (50%)	207
Pb shielding for trunk-line corridor	53

	Annual Costs, $K
316SS, seamless tubing	49
Installation (100%)	49
2 in. Pb shielding from tanks to trunk	53
Valves at 0.15 $K each, one per tank, 296x1x0.15	44
Vacuum pumps of 10 yr life, one for each trunk = 10/10	1
Installation (100%)	1
Monitoring with alarms at 0.4 $K per tank = 296x0.4	118
Installation (100%)	118
Replacement parts for monitoring for 50 years = 296x0.02x50	296
Capital and maintenance costs for loading	1,576+0.296(DW$)
Operating costs (cf. Section 3.1.1.3)	
- during loading	800
- during storage	1,300
Subtotal	2,100
Total cost	3,676+0.296(DW$)

Adding 30% for contingencies brings the total to $4,780+0.385 DW$ for the storage of 169 m^3 (STP) of radiokrypton.

The latter total would be the same as that of subatmoshperic storage in a building (6,450 $K) if the dry well cost roughly $4,300. The actual cost of the dry well would depend upon the medium in which it was emplaced.

3.4.3 Storage in a Deep Geological Medium

3.4.3.1 General Aspects

Experience might prove the surveillance of subatmospheric tanks in a building is unnecessary. In that case one could consider deep geological storage in which a storage room would be sealed very early in the period of storage. Satisfactory media might be basalt, tuff or granite, for example. A mine room in one of these media could serve the same purpose as a cell in Figure 3-9. It would be unnecessarily expensive to store the tanks in dry wells in the room, but mounting the tanks above the floor for convective cooling would be useful. Each FT-size tank would generate a maximum of only 67 W of heat.

Each room would be completely outfitted with mounted tanks and tubing before the room is sealed with the shielded, connecting valves being just outside the seal. The necessary vacuum equipment and trunk lines also would be contained in this gallery outside the storage room, to which the radioactive krypton would be brought in shielded, high-pressure 50-l cylinders. All of the shielded cylinders and even the vacuum equipment could be retrieved from a gallery before the gallery itself was eventually sealed.

3.4.3.2 Environmental Assessment

Geological storage would benefit from the advantage of subatmospheric storage because outward pressure (and presumably outward leakage) would be impossible. There would be negligible cyclic temperature change in a storage room. Even if a tank should have inward leakage, there would be extremely little "breathing" of the tank to permit krypton loss. Geological storage would be immune to tornadoes and hurricanes as well as less vulnerable to sabotage. However, it would be somewhat vulnerable to ingress of water.

3.4.3.3 Estimated Cost

The cost for the disposal of 169 m^3 (STP) of radiokrypton with 296 standard FT tanks of carbon steel is estimated here. The tanks would be mounted on racks to typically give 296 tanks in a room 20 ft high x 78 ft x 78 ft for a thermal loading over the room area of 50 kW/acre. Compare Section 3.4.1.3. A 10 percent premium for underground construction is charged against KSF capital costs and installations.

	Annual Costs, $K
Prior bottling at 500 psi (see Section 3.1.1.3)	47
Loading 500 psi cylinders into casks	15
From capital costs of KSF-type offices, change rooms, etc., 150 ft stack above ground, and head end to unload 500 psi cylinders from casks = (1/30)(5,200) (1.10)	191
Facility for filling FT tanks from 500 psi cylinders	10
Costs of 48 casks of Blomeke-Perona type[13] (70 $K each, 30-yr. life, 3 cylinders cask) = 48x70/30	112
296 FT tanks at 1.4 $K each	414
Installation of tanks (110%)	455
Valves, lines, vacuum pumps, etc. (Section 3.4.1.3)	160
Total operating costs (cf. Section 3.1.1.3)	1,600
Total exclusive of geological costs and transportation	3,004

Geological cost:[14]

	Abandoned Mine	Horizontal Tunnel
From capital costs (Section 3.3.3.3)	65	130
Engineering (5% of latter)	3	7
	68	137
Transportation to		
Eastern US site	37	37
Western US site	99	99
Total costs		
Eastern US site	3,109	3,178
Western US site	3,171	3,240
Adding 30% for contingencies brings the total costs to:		
Eastern US site	4,040	4,130
Western US site	4,120	4,210

These estimates have an uncertainty of ±40%.

3.4.4 Storage in the Ocean

3.4.4.1 General Aspects

A low-pressure tank of the FT type would be unable to withstand the unrelieved pressure at the bottom of the ocean. However, water might be freely admitted as the tank is allowed to sink from the surface to the bottom of the ocean as long as the gas remained captive within the tank. The pressure differential across the tank wall would remain zero. This is a variation of an idea of M. Laser et al and would supposedly lead to the formation of a solid krypton hydrate in the depths of the ocean.[14,15] The tank would initially be evacuated and filled with one-half atmosphere of radioactive krypton at a ship before being dropped to the ocean bottom at a mid-plate/mid-gyre location. The radioactive krypton would be transported to the site in high-pressure cylinders (perhaps 500 psi) in order to minimize shielding and transportation costs.

The operation described above would not require the pressure-activated valves mentioned by Laser, but it would require larger tanks (762 l) and more of them (444) compared with about 145 tanks of 42.5-l size for 500 psi gas.

It would probably be unnecessary to put the tanks in projectiles in order to keep the gas captive. The material of the tank could be the usual carbon steel of limited resistance to sea water. Alternatively the FT tank could be manufactured of nickel or possibly titanuim at half the wall thickness, since the strength and corrosion resistance would probably be adequate. However, these two materials are expensive.

3.4.4.2 Environmental Assessment

It is assumed that the gas could be kept captive by some means until the tank rests on the seafloor. A potential problem might be the eventual loss of krypton by corrosion of the tanks before solid krypton hydrate forms. Most likely, the gas could dissolve completely in the seawater admitted to the tank before the carbon steel is breached by corrosion. However, it is believed that it would require centuries for the abyssal seawater to circulate to the surface of the ocean. This should constitute a satisfactory radiological defense due to the rapid decay of the krypton-85.

3.4.4.3 Estimate of Cost

The cost is estimated below for the disposal of the nominal amount of radiokrypton with 444 standard FT tanks of carbon steel.

	Annual Costs, $K
Bottling room with personnel and maintenance (Section 3.1.1.3)	
145 500 psi cylinders used over 30 yrs = 145x0.5/30	2
Land-based hot cell, loading of shipping casks, costs of 48 casks, concomittant operating expense (Cf. Section 3.1.1.3)	942
444 FT tanks at 1.4 $K each	622
Transportation by land for casks (p. 3-18) and FT tanks	67
Total cost of land operations	1,680
Costs of marine portion (Section 3.1.4.3) without projectiles	952
Total of land-based and marine portions	2,632

The latter is brought to 3,420 $K (±40%) by the addition of 30% for contingencies.

REFERENCES

1. R.A. Brown, et al, eds., "Reference Facility Description for the Recovery of Iodine, Carbon and Krypton from Gaseous Wastes," Report ICP-1126, Idaho Operations Offic, Idaho Falls, Idaho, Apr 1978.

2. R.A. Brown, ibid, p.63.

3. R.B. Bird, W.E. Stewart and E.N. Lightfort, "Transport Phenomena," p.415 (1960).

4. B.A. Foster and D.T. Pence, "An Evaluation of High Pressure Steel Cylinders for Fission Product Noble Gas Storage," Report ICP-1044, Idaho Operations Office, Idaho Falls, Idaho, Appendix A, Feb 1975.

5. B.D. Power, "High Vacuum Pumping Equipment," Reinbhold Publishing Corporation, N.Y. (1966).

6. L. Holland, et al, eds., "Vacuum Manual," E, and F.N. Spon, London (1974).

7. T.R. Pinchback and D.A. Knecht, "The Development of Process and Storage Materials Suitable for Krypton-85 Waste Management," International Symposium on Science Underlying Radioactive Waste Management, Boston, Mass., Nov 29-Dec 1, 1978.

8. B.J. Thamer and P.J. Macbeth, "The Evaluation of Alternatives for the Ultimate Disposal of Krypton-85, Task 2, Comparison of Krypton-85 Storage Techniques," Ford, Bacon and Davis Utah, Salt Lake City, Utah, Mar 1979.

9. U.S. Nuclear Regulatory Commission, Regulatory Guide 1.29, Seismic Design Classification, Revision 2, Feb 1976.

10. U.S. Nuclear Regulatory Commission, "Standards for Protection Against Radiation," Code of Federal Regulations, Title 10, Part 20, Jan 1, 1976.

11. R.A. Brown, ibid, Table 8.

12. R.D. Small and D. Weihs, "Thermal Traces of a Buried Heat Source," Trans. ASME, J. Heat Transfer, p. 47, Feb 1977.

13. J.O. Blomeke and J.J. Perona, "Management of Noble-Gas Fission Product Wastes from Reprocessing Spent Fuels," Report ORNL-TM-2677, Oak Ridge National Laboratory, Oak Ridge, TN, Nov 21, 1969.

14. P. Macbeth, et al, "Evaluations of Alternative Methods for the Disposal of Low-Level Radioactive Wastes," Report

NUREG/CR-0680 to the U.S. Nuclear Regulatory Commission, Low-Level Waste Branch, by Ford, Bacon & Davis Utah, Salt Lake City, UT, April 1979.

15. M. Laser, et al, "Off-Gas Treatment and Krypton Disposal in HTGR-Fuel Element Reprocessing," in OECD/IAEA Symposium on the Management of Radioactive Wastes from Fuel Reprocessing," Paris, 1972.

4. DISCUSSION AND SUMMARY

4.1 DISCUSSION

The most promising techniques for immobilizing krypton-85 have been examined in terms of their salient characteristics, environmental and safety hazards, cost, retrievability, and sociopolitical considerations. Buildings, dry wells, deep geological media, and the ocean have been considered as locations for placing the immobilized radiokrypton. The studies support the following conclusions.

4.1.1 Cost

The cost estimates of this report are shown in Figure 4-1 with uncertainty bands. Since errors in the cost estimation process tend to impact each estimate in the same manner, comparisons are valid.

- The lower costs for disposal at 2000 psi compared with those for 500 psi are due to the reduced space required.
- Storage in the Krypton Storage Facility is expensive because of the high capital and operating costs.
- Ocean disposal is inexpensive because there is no capital cost at the disposal site and the cost of transportation over the ocean is not high. It is assumed that no surveillance is required.
- The storage of krypton alloys from ion implantation/sputtering is generally expensive because of the cost of producing the alloys.

The most expensive option of Figure 4-1 would only add 0.002 percent to a consumer's electric bill of $.05 per kWh. However, the methods of storage of this report would cost $400,000 to $1,200,000 for storage in order to avert a health effect. Comparatively small reprocessing costs to produce the radiokrypton would be extra. While cost is important, safety, reliability, and sociopolitical considerations should also be considered.

4.1.2 Storage of Pressurized Gas

The storage in cylinders of pressurized gas exacerbates radiation-caused corrosion by the presence of impurities other than noble gas. It is assumed that oxygen-, fluorine- and chlorine-containing impurities would be kept below 0.01 percent for this reason. Even so, valved cylinders of pressurized gas probably would require surveillance throughout the periods of

186<

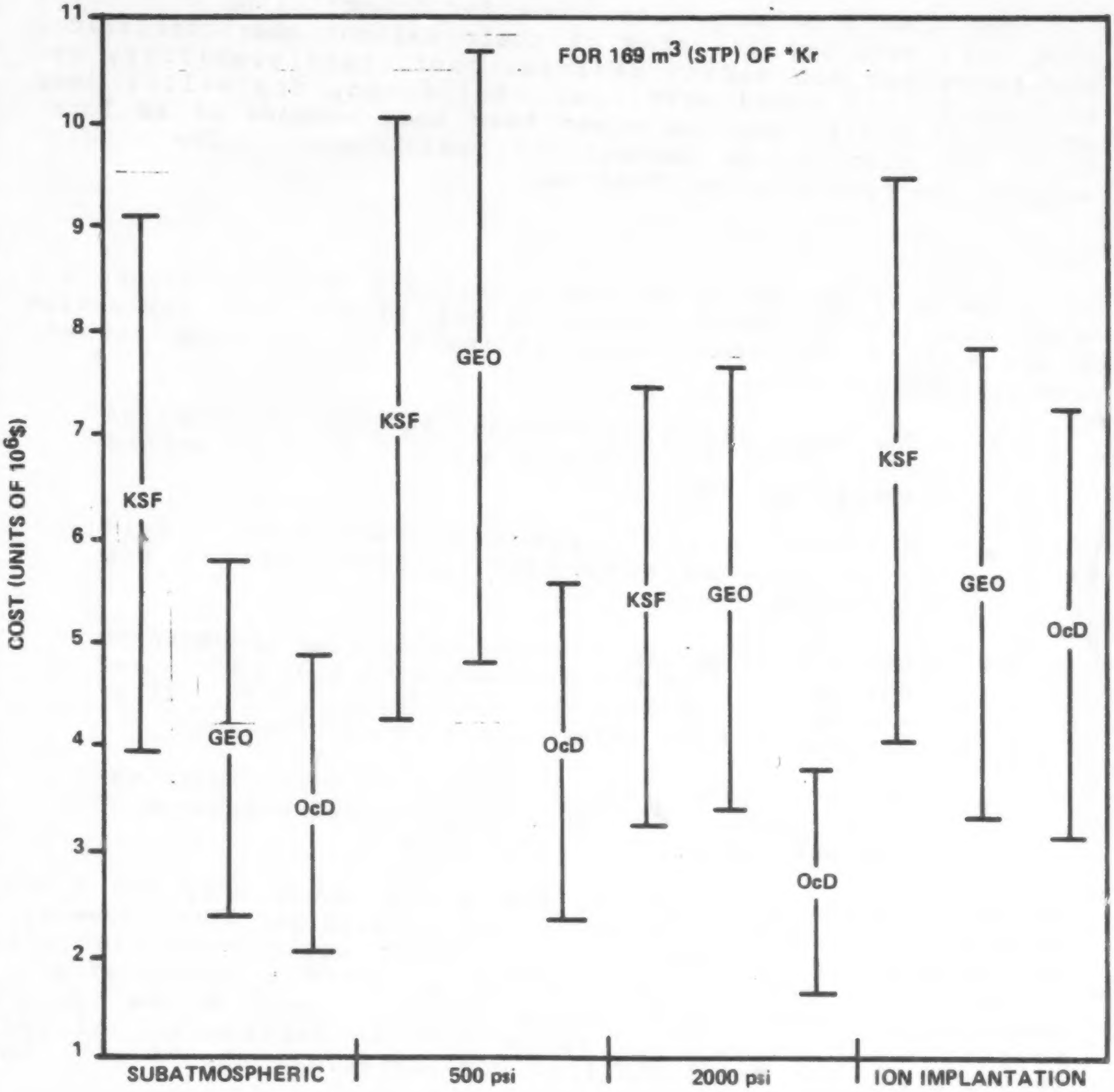


FIGURE 4-1 ESTIMATED COST OF STORAGE FOR KRYPTON - 85

(KSF IS KRYPTON STORAGE FACILITY; GEO IS GEOLOGICAL STORAGE; AND OcD IS OCEAN DISPOSAL) loading and storage because of potential leakage at the valve. toward gaseous krypton. The replacement of valves and threaded joints with welded closures might reduce the leakage, but this option appears not to have been investigated for krypton-85. The leakage of any closure at the sea floor in a mid-plate/mid-gyre location is probably less serious because there is high solubility of the krypton in seawater at such depths and the seawater there requires many tens of half-lives of krypton-85 to circulate to the ocean's surface.

The least expensive immobilization technique for storage in a building appears to be that of 2000 psi gas in a cylinder. Using a cylinder rated for 6000 psi in this case together with spray cooling to back up refrigeration cooling would reduce the chance of a cylinder's suddenly failing. However, the least expensive combination in this study, and one of the more promising overall, is the emplacement, head down, of a 2000 psi cylinder in the seabed. This cylinder and its closure are considered to be made of a metal such as a nickel alloy that resists seawater. However, when leackage occurs, it would very likely be at the closure and the radiokrypton would remain captive within the cylinder as gas or the solid krypton hydrate until it would dissolve in the seawater. As mentioned previously, the abyssal seawater does not communicate with the ocean's surface, for all practical purposes.

4.1.3 Storage of a Krypton Alloy from Ion Implantation/Sputtering

If research with immobilization of krypton alloys should be successful, this storage technique could be far safer than those using pressurized gas. The lack of high temperature or pressure at any time would be an advantage, but cost would remain high.

4.1.4 Storage of Subatmospheric Gas

Subatmospheric storage should be more promising overall than storing pressurized gas in a geological medium. A subatmospheric tank should be comparatively leak-proof and it could be remotely evacuated if leakage should occur. The ability to perform the remedial action remotely recommends the possible use of subatmospheric storage in a geological medium which could be sealed. Subatmospheric storage would have no value in ocean disposal unless pressure equalization were achieved. To equalize pressure the seawater could be admitted to the tank as soon as it began to sink. No specially designed valve would be required to do this.

4.2 SUMMARY

In summary, this report finds the following particularly favorable combinations from among the three main modes of disposition and from techniques of immobilization:

188<

- Storing krypton alloy from ion implantation/sputtering in a building offers a very low emission rate of radioactivity according to present research. Surveillance, remedial action, retrievability, eventual decomissioning, and sociopolitical aspects would be relatively good. However, this combination would be almost as expensive as storing 500 psi cylinders at the same location.

- Storage of subatmospheric tanks of radiokrypton in a deep geological medium could be achieved by remote initial filling via lines to the tanks in a sealed chamber. This would be characterized by low emission rates from the tanks, and remote surveillance and retrieval of the contents of the tanks could be accomplished through the lines. The cost would be two-thirds that of the above combination.

- Storage of 2000 psi radiokrypton in the ocean at a mid-plate/mid-gyre seafloor would be promising if the gas were contained in cylinders of a corrosion-resistant alloy (perhaps nickel-based) and capable of withstanding at least 6000 psi externally. Any gas that escaped would be converted to solid krypton hydrate and/or dissolved in the seawater. Circulation of radiokrypton to the surface of the ocean would be very small. This combination would be the least expensive of all. There would be no retrievability. A detailed study might show that a gaseous pressure higher than 2000 psi would be optimum.

APPENDIX A

COMPARISON OF IMMOBILIZATION ALTERNATIVES IN A SURFACE STRUCTURE

It has been necessary to consistently compare the alternatives of the Task 1 survey[1] in order to select the most promising techniques of immobilization for the present report. The simple system of A (excellent) to F (unacceptable) has been used in categories of Technical Feasibility, Safety, Comparative Cost and Other Factors. The latter includes retrievability, sociopolitical, and transportational aspects. The designation Technical Feasibility is intended to include engineering aspects. The Net Rating is an average of the four equally weighted categories.

A.1 GAS AT 500 PSI IN CYLINDERS

The several advantages of containing the krypton in pressurized steel cylinders have been detailed by B.A. Foster and D.T. Pence.[2] An improved resistance to corrosion for a century might justify the cost of making the cylinder of alloy A286 or some stainless steel, for example. Corrosion studies are in progress at the Idaho National Engineering Laboratory.[1,3] The best seal at the valve seat is currently assumed to be of a metal-to-metal type such as a stem of Type 303 stainless steel and seat of monel.[4] Leakage at the valve (Fig. A-1)[5] can be from (1) the threaded area where the valve is screwed to the cylinder, (2) the plug above and to the right of the valve seat, or (3) some location along the length of the stem. The threads are often covered with teflon tape before joining and this tape-metal combination may have limited durability in the high radiation field. Valves that leak either gradually or suddenly are a potential problem with gas cylinders no matter what the cause of the leakage. The Krypton Storage Facility has been envisioned to have some capability for handling leaky cylinders of krypton-85.[5] M. Laser et al, have estimated a special cylinder valve in a similar facility to leak about 1 x 10^{-8} torr liter/sec.[6] The valve's inlet and the valve's connection to the steel vessel would be welded. The cylinders are considered to be stored at the site of the reprocessing facility when the KSF is used.

This alternative serves as the reference case in Table A-1 and is accordingly rated B in all categories.

A.2 GAS AT 2000 PSI IN CYLINDERS

It is natural to ask what benefits might accrue and what problems might arise if the krypton pressure in the cylinder were increased to 2000 psi since the cylinders are ordinarily certified for this pressure.[4] Cylinders at this pressure could be accommodated on site with appropriate modifications of the Krypton Storage Facility. The storage area would be

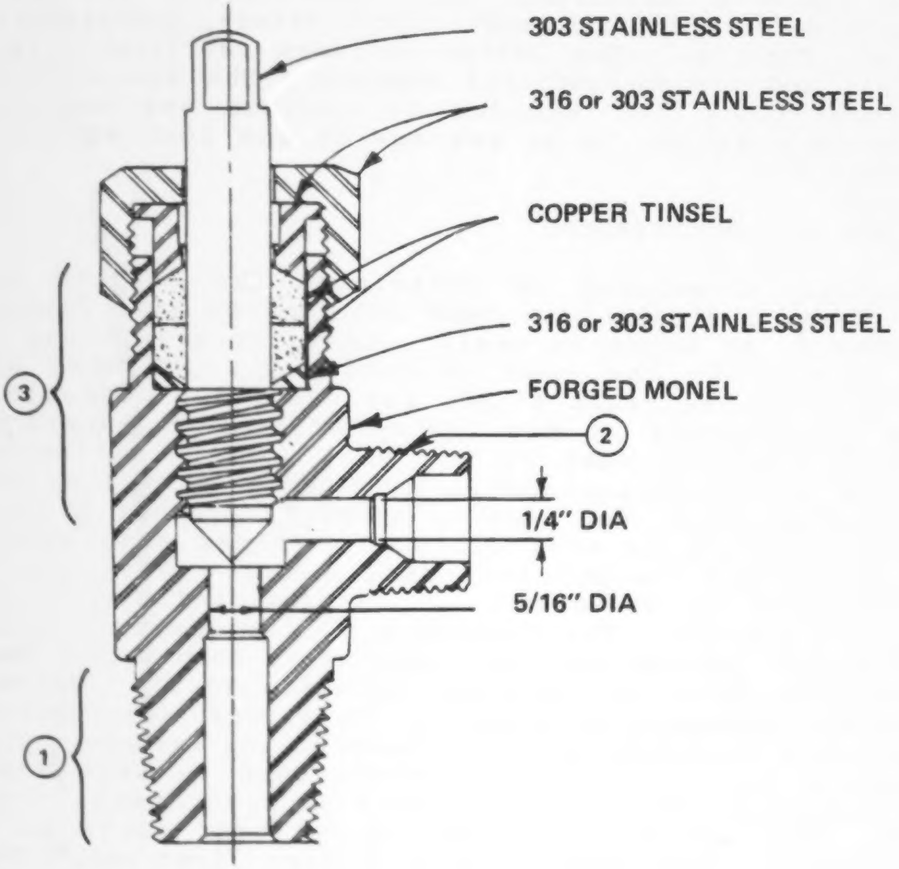

303 STAINLESS STEEL
316 or 303 STAINLESS STEEL
COPPER TINSEL
316 or 303 STAINLESS STEEL
FORGED MONEL
②
1/4" DIA
5/16" DIA
③
①

FIGURE A-1 GAS VALVE

REFER TO P. A-1 FOR ① ② and ③

(SOURCE: REF. 5)

TABLE A-1

SUMMARY OF IMMOBILIZATION TECHNIQUES FOR ^{85}Kr IN A SURFACE STRUCTURE

Immobilization Technique	Technical Feasibility	Safety	Comparative Costs	Other Factors	Net Rating
500 psi Gas Cylinders	B	B	B	B	B
2,000 psi Gas Cylinders					
--with air cooling backup	B	B-	A-	B	B
--with water cooling backup	B	B	A-	B	B to B+
Zeolites	B-	A-	B	B	B
Zeolites with 500 psi of Gas	B-	B+	B+	B	B
Krypton Loaded Solid Set in a Metal Matrix	C+	A-	C	A-	B
Ion Implantation/Sputtering	B	A	B	A-	B+
Metal Vapor Deposition	B	F	C	A-	C to C+
Clathrates	C-	D+	C	A-	C to C+
Porous Underground Media					
--gas	A-	D+	A	C-	B-
--dissolved in solution	A	C	A-	C	B
Hydrofracture					
--entrained gas	B	B-	B+	C	B
--admixed ^{85}Kr-Sodalite	B+	B+	B	C+	B
Extraterrestrial	C+	C+	F	C+	C-
Storage on Charcoal	B	C-	C-	B+	C+

TABLE A-1 (Cont)

Immobilization Technique	Technical Feasibility	Safety	Comparative Costs	Other Factors	Net Rating
Transmutation	D-	C-	F	B	D+
Cryogenic Storage	B+	C+	B	B+	B
Storage in Caverns	B-	C+	A	C	B-
Subatmospheric Storage in Tanks above Ground	A-	A	B	B+	B+ to A-
Solutions	C-	C-	A	B	B-

greatly reduced for handling the same quantity of krypton per year, but the capacity of the air refrigeration per cylinder would have to be improved and the lead shielding should be increased by about 1 inch. These changes could readily be accomplished. However, a failure of the coolant system would have more serious consequences. The nominally 500 psi case under coolant failure has been estimated to result in 192°C and 1000 psi in the cylinder together with 6 psi net pressure across the cell wall.[5] The 2000 psi case could similarly lead to 360°C and 3200 psi in the cylinder and over 12 psi net pressure across the cell wall. This temperature and pressure would be unacceptable except for a cylinder certified for a higher pressure. Likewise the cell wall might not withstand 12 psi across it. Venting would be required. Hence the Safety rating would be less if only ambient air served as the backup for a failure of the coolant system. The safety would improve if there were a fail-safe, emergency cooling system of showers, for example. In this case the Safety rating might be placed at B but no higher because of possible failure of the water coolant system and the generally greater hazard with 2000 psi compared with 500 psi. Either with or without the water-cooling backup the cost would be roughly two-thirds that of the same facility with 500 psi. Except for the above factors, the facility might rate much the same at the two pressures.

A.3 ZEOLITES

Krypton may be loaded into one of the better zeolites like sodalite which would be contained in a porous tube.[7] This enables storage near atmospheric pressure with ordinarily minor leakage of krypton. The latter could easily be contained or recovered for recycling. Hence a Safety rating of A- seems justified for a Krypton Storage Facility although the stability of krypton-loaded sodalite to the radiation of krypton-85 still must be demonstrated. The latter point, poor heat transfer and the required high temperature and pressure of emplacement in the sodalite[8] warrant a comparative Technical Feasibility of B-. The cost would be about the same as that for 500 psi cylinders in similar storage locations. Other Factors might rate B. The public approval of an unpressurized storage system would be diluted by a fear of the high pressures during encapsulation.

A.4 ZEOLITES WITH 500 PSI OF GAS

The inclusion of the radioactive gas at 500 psi with the loaded zeolite (sodalite) would improve the krypton density and reduce the storage space and cost if the problem of thermal loading was not serious.[1] The Technical Feasibility, and to some extent, the Safety and Other Factors would be intermediate between the cases of "500 Psi Cylinder" and "Zeolites."

A.5 KRYPTON-LOADED SOLID SET IN A METAL MATRIX

One of the defects of a zeolite like sodalite as a vehicle for krypton is its poor thermal conductivity in the bulk form. This disadvantage could be removed by suffusing the zeolite with a metal matrix of suitable melting point after the impregnation with krypton.[9] The emplacement of the matrix around the zeolite has some technical difficulties.[9] However, once in place, the matrix could sufficiently improve the heat transfer and resistance to radiation damage to improve the Safety. The added complexity and labor of the process as well as the small added cost of the metal matrix would make matrix alternatives more expensive. The use of carbonyl nickel in place of a metal matrix might warrant some thought, but its use is untested in this application.[1]

A.6 ION IMPLANTATION/SPUTTERING

Ion implantation/sputtering to produce 9.0 atomic percent krypton in an amorphous alloy like 87 at% Fe-13 at%Y is assumed to be attainable.[1] The resultant krypton density of about 174 cm^3 (STP)/cm^3 alloy would be much higher than that of 2000 psi gas even with a krypton-free coating of nickel over the alloy strip. The heat transfer would be very good so that storage in a Krypton Storage Facility could give better economy of space than would 2000 psi gas cylinders. This saving would partly compensate for the cost of the implantation/sputtering itself for a Krypton Storage Facility. Overall cost would be comparable to that of 500 psi cylinders in the Krypton Storage Facility. Technical Feasibility appears to be very good if the storage temperature does not exceed 200° to 300°C and the alloy is sufficiently resistant to radiation. Then Safety should be excellent because of low pressures, temperatures and leakage during both ion implantation/sputtering and storage. The nickel coating could impart good corrosion resistance to the metal. The ratings for Other Factors might correspond to those for zeolites.

A.7 METAL VAPOR DEPOSITION

The option of metal vapor deposition is less promising than that of ion implantation/sputtering in most respects except possibly Other Factors.[1] The attainable krypton density and leakage rate are more than an order of magnitude worse so that Safety and Costs (per cm^3 (STP) Kr) are worse. Otherwise the process of producing this so-called "kryptonate" and capping it is technically feasible.

A.8 CLATHRATES

The Technical Feasibility and Safety of using a clathrate such as that of quinol are generally poor because of its poor thermal stability and conductivity. Only a light loading is feasible and that would be expensive because of the cost of quinol, the large storage space and a need to artificially irradiate the clathrate. Hence the cost would be somewhat worse than that for zeolites, but Other Factors would be similar.

A.9 POROUS UNDERGROUND MEDIA

Krypton-85 in a fluid form could be injected into a porous geological medium covered by caprock.[1] The Technical Feasibility of this appears to be high provided the site is suitably selected. However, low leakage rates have not been demonstrated as yet with gas although they should be achievable. A solution in a perfluorinated hydrocarbon for example, could sink quite low and never give up all of its krypton to pores in the soil. The cost with the use of gas would be low as pointed out by P. C. Reist,[11] and that for a liquid solvent would depend upon the solvent. Other Factors would not rate highly because retrievability would be impossible, transportation might be required to suitable sites, and there might be public disapproval of even small amounts of krypton-85 rising from the ground.

A.10 HYDROFRACTURE

Krypton gas could likely be entrained in grout for hydrofracture in a facility such as that at Oak Ridge National Laboratory. The most important barrier against this krypton reaching the atmosphere and the public would be the medium at the fracture, e.g., clay-like shale, and hundreds of meters of depth. The safety would be better than that of Porous Underground media. The cost would be shared with that of the hydrofracture disposal of other radioactive waste and should not be high. Other Factors might be only slightly better than it is with Porous Underground Media for similar reasons. The Technical Feasibility and Safety would be improved if the krypton were entrained with the grout in the form of krypton-loaded solid such as sodalite. The expense would be increased by the loading of the krypton into the sodalite.

A.11 EXTRATERRESTRIAL

Extraterrestrial disposal should be achievable with a large program of development.[1] However, the possibility of an accident at launch or shortly thereafter is too strong to be ignored. Costs would be prohibitive, particularly with protection against an abort or reentry. Retrievability would be extremely difficult and public acceptance would be low in spite of the potential to completely remove the radioactivity from the earth.

A.12 STORAGE ON CHARCOAL

The alternative of storage on adsorbents is best exemplified with charcoal. Charcoal has been much used for the (temporary) storage of radio-xenon and -krypton; thus, the Technical Feasibility appears to be good. However, the krypton is not tightly held on charcoal and could fairly easily be driven off by moisture or gas, particularly by a fire on this readily burnable adsorbent. Hence, charcoal's safety is no better than fair. One would need large amounts of charcoal, large containers and large amounts of shielding. Hence, charcoal would be fairly expensive. The rating for Other Factors might be like that of zeolites except that the public might dislike the flammability of charcoal.

A.13 TRANSMUTATION

The Technical Feasibility of transmutation is very poor in any practical sense at the present time. The separation of krypton isotopes, transportation to an accelerator for spallation neutrons, and long irradiations would make this an expensive process. The engineering of suitable safety during irradiation might be difficult because of radiation effects and the need for thin windows and high thermal stresses. There would be public approval for removing krypton-85 rather than storing it. However, the required transportation might introduce political problems.

A.14 CRYOGENIC STORAGE

Cryogenic storage would likely be on the site where the radioactive gas would be produced. The krypton might be stored as solid krypton or a solid krypton-xenon mixture cooled with liquid nitrogen at about -180° to -190°C. Alternatively the radioactive gas might be dissolved in liquid nitrogen in a closed system that is externally cooled, probably by liquid nitrogen. Although the routine cost of coolant would be small, one would have to have a reliable source of it or other cooling. A large, diesel-powered plant for liquid nitrogen might be located on the site. However, in case of coolant failure the krypton could be vented into an inexpensive storage tank for recovery or, as a last resort, vented through the stack of the reprocessing plant. The compactness of the condensed krypton would lead to savings in shielding and building space. Overall Costs would rate B. The feasibility of cryogenic storage is partly demonstrated in the cryogenic processes for producing krypton-85.[1] However, if the radio-krypton is to be contained in metal, the impurities other than xenon should be considerably reduced. Hot-trapping the krypton before storage might accomplish this. This would also combat the hazard of an explosion between condensed hydrocarbons and oxygen or ozone, which also must be dealt with in the liquid nitrogen. Safety might rate C+. Other Factors might rate B+ because of excellent retrievability, no transportation problems for the krypton, probable public approval of a low-pressure system, but disapproval of a possible explosion.

A.15 LOW-PRESSURE STORAGE IN CAVERNS

A suitably closed cavern might be loaded with radioactive krypton in an amount that would be a small fraction of the volume of the cavern. Thus the pressure to produce leakage

197<

would be small and any leaks would be highly diluted. Furthermore, leakage could be controlled when it occurred by exhausting some of the cavern's contents through a stack. The process should be quite inexpensive. The retrievability would be rather poor. Political and social problems might arise from the need to transport radioactive krypton to a suitable cavern.

A.16 LOW-PRESSURE STORAGE IN TANKS

The Operations Division of Oak Ridge National Laboratory has for years dispensed krypton-85 to customers in tanks of less than one atmosphere of absolute pressure.

The tank would be evacuated before filling to less than atmospheric pressure with purified gas. Even with the decay heat from the dilute gas, the pressure within the tank would be less than atmospheric so that any leakage would tend to be inward. The dilute gas would make any gas corrosion and radiation effects much less serious than they would be for 500 to 2000 psi gas in a cylinder. The leakage should be much less serious than with 500 psi cylinders. However, a leaking tank could be detected by its rising pressure and with radiation-monitoring equipment. Evacuation could be used either to recover the contents of leaking tanks or to prepare the facility for decommissioning. The above indicates a high degree of Safety and Technical Feasibility. The cost is estimated to be no more than that of 500 psi gas cylinders. The public might more readily accept subatmospheric tanks than 500 psi tanks because of manifestly less chance of outward leakage. Hence other factors might rate B+.

A.17 SOLUTIONS

The best use of krypton dissolved in perfluorinated organic compounds is probably in Porous Underground Media (paragraph A.9) because of the corrosiveness of its radiolytic products. Solutions in polyphenyls would have radiolytic products that would be noncorrosive but flammable if evolved hydrogen were allowed to concentrate. One terphenyl mixture melts in the range of 50° to 145°C. This material could possibly be buried as a solid after high pressure impregnation as a liquid. This subject has not been researched. Its Technical Feasibility and Safety would hinge on its low evolution of krypton in the presence of air, water or soil. Hence these categories might be rated C- at present. The cost of terphenyl and burial would probably be less than 20% of that of 500 psi cylinders in the Krypton Storage Facility. Other Factors would be favored by good retrievability, probable storage at the site of the reprocessing plant, and no public distrust of high pressure storage.

Dissolving krypton in glass may be developed into a suitable means of storage. The Battelle Pacific Northwest Laboratory is pursuing this subject.[12]

198

A.18 COMBINATIONS OF ALTERNATIVES

The preceding sections have included certain synergistic combinations such as loaded zeolite admixed with grout in hydrofracture (paragraph A.10) and pressurizing a container of loaded zeolite with the radioactive gas (paragraph A.4).

REFERENCES FOR APPENDIX A

1. B.J. Thamer et al, "The Evaluation of Alternatives for the Ultimate Disposal of Krypton-85, Task 1 - A Compilation of Alternatives," Report UC-261-01, Ford, Bacon and Davis Utah, Inc., Salt Lake City, Utah, Feb, 1979.

2. B.A. Foster and D.T. Pence, "An Evaluation of High-Pressure Steel Cylinders for Fission Product Noble Gas Storage," Report ICP-1044, Idaho Operations Office, Idaho Falls, Idaho, Feb 1975.

3. T.R. Pinchback and D.A. Knecht, "The Development of Process and Storage Materials Suitable for Krypton-85 Waste Management," paper presented at the International Symposium on Science Underlying Radioactive Waste Management, Boston, Mass., Nov 29-Dec 1, 1978.

4. C.W. Nielsen, "Safety Analysis Report for Packaging Krypton, Shipping Container USA/5958/BL (ERDA-ID)," Report ICP-1077, Idaho National Engineering Laboratory, Idaho Falls, Idaho, June 1975.

5. R.A. Brown et al, eds., "Reference Facility Description for the Recovery of Iodine, Carbon and Krypton from Gaseous Wastes," Report ICP-1126, Idaho Operations Office, Idaho Falls, Idaho, April, 1978.

6. M. Laser, Sections "Type of Containers," and "Valves" in "Separation, Storage and Disposal of Krypton-85," Working Paper prepared for a Technical Committee of the International Atomic Energy Agency, Vienna, Austria, 1979.

7. R.W. Benedict, "An Evaluation of Methods for Immobilizing Solids Loaded with Krypton-85," Report ICP-1130, Allied Chemical Corporation, Idaho Falls, Idaho, Aug 1977.

8. D.A. Knecht, "An Evaluation of Methods for Immobilizing Krypton-85," Report ICP-1125, Idaho National Engineering Laboratory, July 1977.

9. K.M. Lamb, "Final Report: Development of a Metal Matrix for Incorporating High-Level Commercial Waste," Report ICP-1144, Allied Chemical Corp., Idaho Falls, Idaho, March 1978.

10. G.L. Tingey et al, "Entrapment of Krypton in Sputter Deposited Metals - A Storage Medium for Radioactive Gases," Report PNL-2879, Battelle Pacific Northwest Laboratory, Richland, Wash., 1979.

200

REFERENCES FOR APPENDIX A (Cont)

11. P.C. Reist, "The Disposal of Radioactive Krypton-85 in Porous Media," Report NYO-841-7, Department of Industrial Hygiene School of Public Health, Harvard University, 665 Huntington Ave., Boston, Mass., Oct 26, 1965.

12. W.J. Gray and G.L. Tingey, "Krypton Dissolution in Low Density Glass," of A.M. Platt and J.A. Powell (comp.), "Nuclear Waste Management Quarterly Progress Report, January through March, 1978," Report PNL-2378-1, National Technical Information Service, U.S. Department of Commerce, Springfield, VA., Aug 1978.

APPENDIX B

CALCULATIONS OF RADIOACTIVITY AND REFRIGERATION

The location of the emplacement of the stored krypton-85 may be called a "facility" if the radiokrypton is to be readily retrievable as in a building such as the Krypton Storage Facility, a dry well, or a geological cavern. The schedule of loading, storage and removal of Section 2.1.3 should apply in these cases. Ocean disposal does not ordinarily feature retrieval. Hence, the above schedule would be inapplicable in this case, beginning with the period of removal. Likewise the dissipation of radioactive heat must be considered in a facility, but is not a serious problem in deep ocean disposal.

B.1 RADIOACTIVITY

The facility is assumed to be loaded at a constant rate L (Ci/yr) and to undergo a constant fractional loss of radioactivity c per year due to operational losses during the loading period t_L. The losses are assumed to be through a large stack. The applicable differential equation is

$$dN/dt = L-(\lambda+c)N \tag{B.1}$$

where N is the number of curies in the facility and $\lambda = (\ln 2)/10.73 = 0.064{,}599/\text{yr}$.

The integration of Equation B.1 yields

$$N = (L/(\lambda+c))(1-e^{-(\lambda+c)t}) \tag{B.2}$$

The world public is exposed to leaked radioactivity N_{LL} during the loading period:

$$dN_{LL}/dt = cN \tag{B.3}$$

Combining Equations B.2 and B.3, and integrating, one obtains

$$N_{LL} = (cL/(\lambda+c))[t-(1/(\lambda+c)](1-e^{-(\lambda+c)t}) \tag{B.4}$$

and $t = t_L$ at the end of the loading period.

During the period of simple storage following the loading period,

$$dN/dt = -(\lambda+c)N \tag{B.5}$$

Combining equation B.2 with $t=t_L$ and the integrated form of Equation B.5 gives

$$N = (L/(\lambda+c))(1-e^{-(\lambda+c)t_L})e^{-(\lambda+c)t} \tag{B.6}$$

The world public is exposed to leaked radioactivity N_{LS} during the storage period:

$$dN_{LS}/dt = cN \tag{B.7}$$

Combining Equations B.6 and B.7, and integrating gives

$$N_{LS} = (cL/(\lambda+c)^2)[1-e^{-(\lambda+c)t_L}](1-e^{-(\lambda+c)t}) \tag{B.8}$$

where $t = t_s$ at the end of the storage period.

As the containers are removed and vented through a stack, the world public is thereby exposed to removed radioactivity N_R such that

$$N_R = Lte^{-(\lambda+c)(t_L+t_s)} \tag{B.9}$$

and t becomes t_L as the last cylinder is vented. During this period the radioactivity N remaining in the facility is determined by

$$dN/dt = -Le^{-(\lambda+c)(t_L+t_s)} -(\lambda+c)N \tag{B.10}$$

Integrating the latter from t = 0 at the onset of the removal period gives

$$\begin{aligned} N = {} & (L/(\lambda+c))[1-e^{-(\lambda+c)t_L}]e^{-(\lambda+c)(t+t_s)} \\ & -(L/(\lambda+c))[1-e^{-(\lambda+c)t}]e^{-(\lambda+c)(t_L+t_s)} \end{aligned} \tag{B.11}$$

where N at t=0 is that from Equation B.6 with $t = t_s$. Of course, Equation B.11 gives N = 0 at $t = t_L$, i.e., when the facility is empty. The curve of Figure B-1 has been calculated using the schedule of Section 2.1.3 with L = 15×10^6 Ci/yr,(1-2) c = 0, and Equation B.2 during loading, Equation B.6 during storage, and Equation B.11 during the removal of containers.

During the removal period, the radioactivity N_{LR} leaking from the facility is determined by

$$dN_{LR}/dt = cN \tag{B.12}$$

where N is from Equation B.11. Performing the integration gives

$$\begin{aligned} N_{LR} = {} & (cL/(\lambda+c)^2)e^{-(\lambda+c)t_s}[1-(1+t/(\lambda+c))e^{-(\lambda+c)t_L}] \\ & -(cL/(\lambda+c)^2)e^{-(\lambda+c)t_s}[e^{-(\lambda+c)t}(1-e^{-(\lambda+c)t_L}) \\ & -e^{-(\lambda+c)t_L}(1-e^{-(\lambda+c)t})] \end{aligned} \tag{B.13}$$

where $t = t_L$ at the end of the removal period.

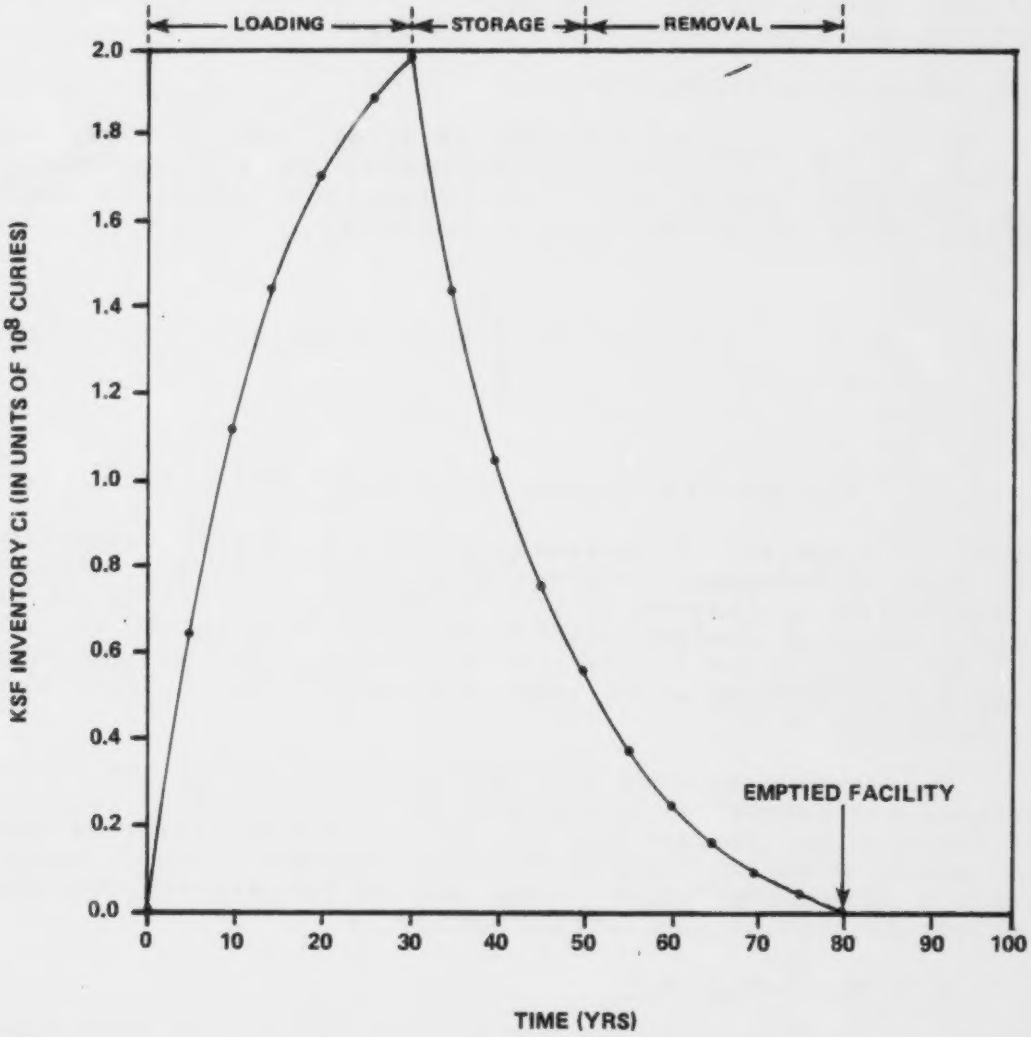


FIGURE B-1 INVENTORY OF THE KRYPTON STORAGE FACILITY WITH L = 15×10^6 Ci/YR AND c = 0.

The cumulative emissions are plotted in Figure B-2 using Equation B.4 during loading, Equation B.8 during storage, and Equations B.9 and B.13 during the removal of containers. The removed containers are assumed to be vented. The annual leakage rate (c) is assumed to be zero, 0.1 percent,[1-2] and 1 percent for the three curves. Table B-1 summarizes the estimated radiological impact upon the world's population.

B.2 REQUIRED REFRIGERATION

The power required for refrigeration, (kW) or \$/yr, during loading is approximately proportional to the inventory N. The aggregate energy E for refrigeration (kWyr) or cost of refrigeration (\$) during loading would be

$$E = (L/(\lambda+c))\int_{0}^{t_L}[1-e^{-(\lambda+c)t}]dt \qquad (B.14)$$

or

$$E = (L/(\lambda+c))[t_L-(1/(\lambda+c))(1-e^{-(\lambda+c)t_L})] \qquad (B.15)$$

where L is the annual increase, (kW/yr) or $\$/yr^2$, respectively, for cooling undecayed radioactivity (15×10^6 Ci/yr). Values of L are given in Chapter 3 in the sections on estimated cost. Thirty years of loading would then give: E = 259.3L for c = 0; 257.4L for $c = 10^{-3}$; and 241.6L for $c = 10^{-2}$. An average year during loading would require 8.64L(c = 0), 8.58L($c = 10^{-3}$), or 8.05L($c = 10^{-2}$).

The energy required for refrigeration during the period of storage and removal of containers can be calculated. During any of the loading, storage and removal periods the ratio of decayed to leaked atoms of krypton-85 is a constant (λ/c). Hence the ratio R of refrigeration energy during the storage and removal periods to that of the loading period is

$$R = (N_{LS} + N_{LR})/N_{LL}$$

where N_{LS} is from Equation B.8 with $t = t_S$, N_{LR} is from Equation B.13 with $t = t_L$, and N_{LL} is from Equation B.4 with $t = t_L$. With the foregoing assumptions it is found that R = 0.747 for c = 0, R = 0.710 for $c = 10^{-3}$, and R = 0.625 for $c = 10^{-2}$. Combining these figures with those of the previous paragraph, the average cost of energy of refrigeration for the sum of the periods of storage plus removal of containers would be 6.45L for c = 0, 6.09L for $c = 10^{-3}$, and 5.03L for $c = 10^{-2}$. These figures are for 169 m^3 (STP) of radiokrypton.

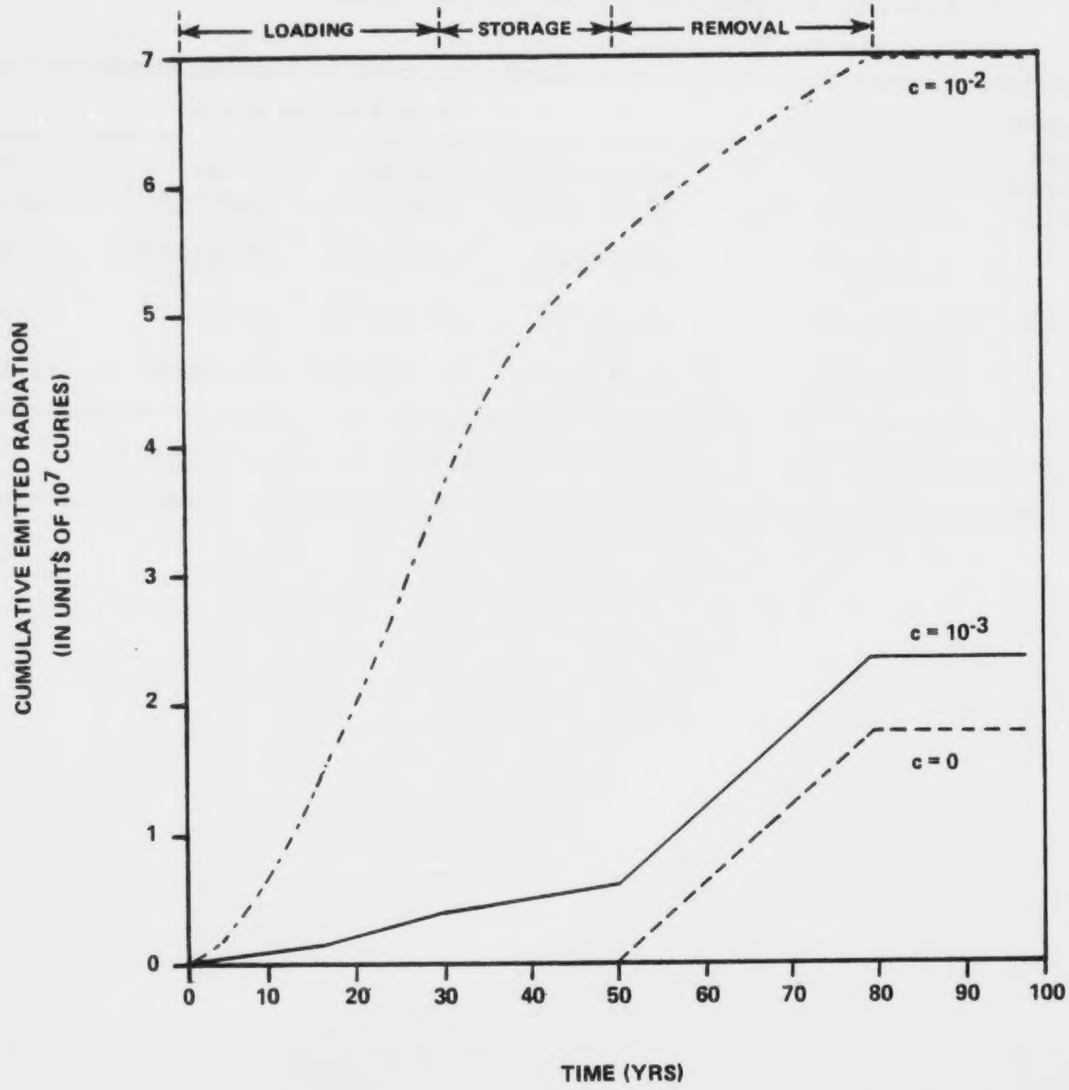


FIGURE B-2 CUMULATIVE EMISSIONS OF KRYPTON - 85 FROM KSF FOR c = 0 (- - -), c = 10^{-3} (——), AND c = 10^{-2} (— - —).

TABLE B-1

FINAL RADIOLOGICAL IMPACT ON WORLD'S POPULATION OF THIRTY YEARS OF LOADING 15,000,000 Ci Kr-85 PER YEAR INTO THE KSF*

Assumed Annual Leakage Rate	Cumulative Radioactivity Exposed to World (Ci)	Population Dose			
		Whole Body (man-rem)	Gonads (man-rem)	Lungs (man-rem)	Health Effects
$c=0$	1.78×10^7	10.4×10^3	13.9×10^3	20.8×10^3	7.8
$c=10^{-3}$	2.35×10^7	13.8×10^3	18.4×10^3	27.6×10^3	10.3
$c=10^{-2}$	6.97×10^7	40.8×10^3	54.5×10^3	81.8×10^3	30.5

*Estimated for a world population of 4.3 billion persons.[3]

APPENDIX B REFERENCES

1. "Environmental Impact Statement: Management of Commercially Generated Radioactive Waste," Report DOE/EIS-0046-D, Department of Energy, Washington, DC, April 1979.

2. "Technology for Commercial Radioactive Waste Management," Report DOE/ET-0028, Department of Energy, Washington, DC, 1979.

3. B.J. Thamer, et al, "The Evaluation of Alternatives for the Ultimate Disposal of Krypton-85, Task 1 - Compilation of Alternatives," Report UC-261-01, prepared for the U.S. Environmental Protection Agency by Ford, Bacon & Davis Utah Inc., Feb 1979.

208<

APPENDIX C

CALCULATIONS OF TEMPERATURE IN THE IMPLANTED ALLOY

The peaking of temperature in the deposited alloy due to the distributed source q (W/cm^3) may be sufficiently estimated by considering an infinite cylinder of the same inside radius (a) and outside radius (b) (in cm). The relevant equation for cylindrical symmetry and the steady state is

$$d^2T/d^2r + (1/r)(dT/dr) + q/k = 0$$

where T = temperature (°K), r = radius (cm), and k = thermal conductivity (W/cm-°K).[1] The solution of the differential equation is of the form

$$T = -qr^2/4k + \mathrm{Clnr} + C_2 \qquad \text{(C.1)}$$

where C_1 and C_2 are constants to be determined. The boundary conditions are $T = T_1$ at $r = a$ and $T = T_2$ at $r = b$. These conditions applied to Equation C.1 allow the determination of C_1 and C_2 and yield the solution:

(C.2)

$$T-T_1 = [(\ln(r/a))/\ln(b/a)][T_2-T_1 + (q/4k)(b^2-a^2)] - (q/4k)(r^2-a^2)$$

40,000 Ci is contained in 1680 g of alloy the density of which may be taken as 5.8 g/cm^3. Thus the initial distributed heat source may be taken as q = (40,000/128,000)(44.5)(5.8/1680) x4.186 = 0.20 W/cm^3 with the aid of figures from B.A. Foster and D.T. Pence.[2] The inside radius a is 7.22 cm and the alloy's outside radius b is 7.78 cm.[3] Equation C.2 is solved for $T_2 = T_1$, a condition which could be approximated in practice, and r at the midpoint between the inner and outer radius. The result is $T-T_1 = 0.007{,}841/k$. The maximum temperature peaking in the alloy would be only 0.8 °C if k for the alloy were as small as 10^{-2} W/cm-°K, a value only 1 percent of that of iron or copper.

REFERENCES FOR APPENDIX C

1. See, for example, B. Gebhart, "Heat Transfer," 2nd. ed., McGraw-Hill Book Co., N.Y. (1971).

2. B.A. Foster and D.T. Pence, "An Evaluation of High Pressure Steel Cylinders for Fission Product Noble Gas Storage," Report ICP-1044, Idaho Operations Office, Feb 1975.

3. G.L. Tingey, et al,. "Entrapment of Krypton in Sputter Deposited Metals, A Storage Medium for Radioactive Gases," Report PNL-2879, Pacific Northwest Laboratory, Richland, Wash., Jan 1979.

210<

GLOSSARY

Term	Definition
1 atm	14.7 psi
at%	atomic percent
1 Ci	3.7×10^{10} disintegrations/second
half life	the time ($T_{1/2}$) required by an isotope to decay to one-half of its previous value; $T_{1/2} = (\ln 2)/\lambda$
health effect	cancer induction and/or mortality
kW	kilowatt
^{85}Kr	the isotope of krypton having an atomic weight of 85 and a half-life of 10.73 years
KSF	The Krytpon Storage Facility, a building described in Report ICP-1126 for storing pressurized gas cylinders of ^{85}Kr.
λ	the decay constant in radioactive decay: $dN/dt = - N$; $\lambda = (\ln 2)/T_{1/2}$
m	meters
MCDF	Mined Cavity Disposal Facility
mrem	10^{-3} rem
MTHM	metric tonnes of heavy metal
MWD	megawatt days of energy
MWe	electrical megawatts
psi	pounds per square inch
psig	pounds per square inch according to the pressure gauge

rem	a measure of the dose of any ionizing radiation to body tissue in terms of its estimated biological effect relative to a dose of 1 roentgen of X-rays
STP	Standard Temperature (0^oC) and pressure (1 atm) for a gas
$K	Thousands of dollars